PB-215 400

INTERIM PROGRESS REPORT ON THE DECONTAMINATION OF RADIOACTIVE WATERS

Roy J. Morton, et al

Oak Ridge National Laboratory
Oak Ridge, Tennessee

May 1953

DISTRIBUTED BY:

NTIS
National Technical Information Service
U. S. DEPARTMENT OF COMMERCE
5285 Port Royal Road, Springfield Va. 22151

PB 215 400

INTERIM PROGRESS REPORT
ON THE
DECONTAMINATION OF RADIOACTIVE WATERS

Prepared for

THE ADVISORY BOARD
FOR RESEARCH ON
DECONTAMINATION OF RADIOACTIVE WATERS

By

Radioactive Liquid Waste Research and Development Section
Health Physics Division
Oak Ridge National Laboratory

May, 1953

Reproduced by
NATIONAL TECHNICAL
INFORMATION SERVICE
U S Department of Commerce
Springfield VA 22151

137

iA

DRAFT MEMORANDUM May 18, 1953

TO: Members of the Advisory Board and Others Concerned.

FROM: Roy J. Morton, Leader of Radioactive Liquid Waste Research and Development Section, HP Division, ORNL, and Conrad P. Straub, Senior Sanitary Engineer, EHC, PHS, Oak Ridge, Tennessee.

SUBJECT: Interim Progress Report on the Decontamination of Radioactive Waters.

Attached hereto is a draft copy of the subject report which is being sent to each member of the Advisory Board for Research on Decontamination of Radioactive Waters.

The ~~With this~~ report there has been included a short summary entitled "Conclusions Based Upon the Interim Progress Report on the Decontamination of Radioactive Waters." It will be found immediately following the title page.

The detailed report which follows the summary covers the following subjects: Research on the decontamination of radioactive waters, the problem of radioactivity in water courses, concentration of radioactivity by natural agents, water decontamination, and instrumentation.

Each section is considered as a unit and the pagination is complete for that unit. All tables and figures carry reference section numbers. This was done to permit amplification or revision of each section or addition of other sections as further information becomes available, and to simplify keeping the report up to date.

Reproduced from
best available copy.

May 18, 1953

It will be noted that graphical-type reporting of results have not been utilized. Where the use of graphs or photographs will aid in depicting certain information, it is hoped that such can be included when the report is reproduced in final form.

In reviewing this report it is hoped that suggestions will be made for improving or expanding this material.

Roy J. Morton
Roy J. Morton

Conrad P. Straub
Conrad P. Straub

CONCLUSIONS BASED UPON THE INTERIM PROGRESS REPORT ON THE DECONTAMINATION OF RADIOACTIVE WATERS

At the October 6, 1950, meeting of the Advisory Board for Research on Decontamination of Radioactive Waters, the Board recommended that research priority be given to:

(1) Evaluation of military type rapid coagulation and filtration apparatus in removing mixed fission products, and

(2) Evaluation of standard water treatment processes and variations thereof in the removal of mixed fission products and individual radioactive isotopes; and as a supplement to this, evaluation of processes suitable for small-scale field decontamination of water supplies.

The Board further recommended that concurrent attention be given to the problems of

(1) Suitable instruments for continuous monitoring of water supplies for the detection of low level radioactive contamination, and

(2) Adequate methods and standards for detection of radioactive contaminants in water under emergency conditions.

These points are discussed in considerable detail in the Interim Progress Report on Decontamination of Radioactive Waters, but will be reviewed here to give a general idea of the progress made in satisfying the recommendations of the Advisory Board.

Since the results pertaining to the evaluation of the military type rapid coagulation and filtration apparatus in removing mixed fission products are classified, they are not included in this discussion. However, for anyone interested, these results will be found in a series of reports prepared by the Engineer Research and Development Laboratories, Fort Belvoir, Virginia.*

Considerable experimental work has been carried out in the removal of radioactive materials from Oak Ridge tap water solutions by conventional water treatment processes. These processes included coagulation and sedimentation, filtration through various media, ion exchange, and lime-soda ash softening. In addition such processes as phosphate coagulation, addition of clay, excess lime-soda ash treatment, and addition of metallic dusts were investigated.

To evaluate the efficacy of any treatment process in a given situation, the concentration of radioactive materials in the contaminated water and the maximum permissible concentration (MPC) of contaminants in the water should be determined as soon as practical. As a preliminary step, the selection of an MPC value would be predicated on the length of time the water was to be consumed. For example, under emergency conditions where short time use of the water is contemplated, the MPC value selected would differ markedly from that selected for a water to be used throughout one's lifetime. In the event that a water is contaminated by a nuclear bomb blast, additional refinements in the selection of an MPC value may be made depending on the age of the fission products mixture. As the final step, regardless of the source of contamination, the radiochemical composition of the water should be determined as time permits. On the basis of this analysis, the MPC value can be defined more closely, and it will also be possible to determine which radioisotopes should be removed.

---

*

1. Lacy, William J. and Don C. Lindsten. Purification of Water Contaminated with Radioactive Materials. Report on Project 8-75-05-008 submitted to Chief of Engineers, U.S. Army. (1 July 1952).

2. Lacy, William J. and Don C. Lindsten. Removal of Radioactive Materials from Contaminated Water by Thermocompression Distillation. Interim Report on Project 8-75-05-008 submitted to Chief of Engineers, U.S. Army. (31 December 1952).

3. Lacy, William J. and Don C. Lindsten. Ion Exchange Materials for Radioactive Water Decontamination. Interim Report on Project 8-75-05-008 submitted to Chief of Engineers, U.S. Army. (1 May 1953).

If a source of water supply suffers a direct hit, the concentration of radioactive materials resulting from the fission products released during the blast may be sufficiently high so as to preclude the production of a safe water supply by the usual water treatment processes. It is only when the water supply is slightly contaminated (concentration of activity in the order of one microcurie/cc or less - 1 μc/cc) that it may be possible, under certain conditions, to produce a satisfactory water by means of existing water treatment plant facilities. A considerable amount of work has been done on the evaluation of conventional water treatment processes or various feasible modifications thereof in the removal of mixed radioactive materials and specific radioisotopes from water. A summary of the data is presented below under various headings.

Removal of Mixed Fission Products

a. Coagulation and sedimentation. - Removals of 45 to 85 per cent were obtained depending upon the composition of the waste and the amounts and kinds of coagulating chemicals employed. Filtration may increase removals a little, as for example, removals of 46 per cent by coagulation and settling were increased to 70 per cent with filtration.

b. Lime-soda ash softening. - Removal efficiencies may be increased to about 90 to 95 per cent with excess treatment.

c. Phosphate coagulation. - Removals of 99.4 to 99.6 per cent have been reported, but these removals were obtained under carefully controlled conditions.

d. Ion exchange. - Where both cation and anion exchange resins have been employed, removals in excess of 99.9 per cent have been obtained. However, the capacity of such units is dependent entirely upon the chemical composition of the water passing through the unit. In general, removals of radioactive materials parallel hardness removal, although some removal (50 to 75 per cent) of the radioactive fraction may continue even though the bed has been exhausted from the standpoint of hardness removal.

e. In all probability, slightly higher removals may be obtained if several processes are combined, particularly if each is specifically suited for certain groups of radioactive materials. In other words, the removals by a combination of different processes may be additive.

Whether the removals indicated above are adequate or not will depend upon the initial concentration of radioactive materials in the water under treatment and the particular MPC value selected. In general, concentrations above 1 μc/cc cannot be reduced to the permissible emergency levels (9×10^{-2} μc/cc for 10 days use immediately after the bomb blast) by coagulation, settling, and filtration alone; other processes such as water softening, phosphate coagulation, or ion exchange will have to be utilized.

Removal of Individual Radioisotopes

Where one is concerned with the removal of specific radioisotopes the method of choice will depend on the required removal efficiency. Under specific test conditions, various individual isotopes have been studied in the laboratory, and whether the resulting removal efficiencies are applicable to other treatment conditions will depend primarily upon the chemical nature of the water with regard to both radioactive and stable elements present. Brief statements concerning the removal efficiencies obtained with individual radioisotopes by various processes under the test conditions reported in the body of the Interim Report are summarized below:

a. Coagulation and sedimentation. - Removals for iodine and cobalt radioisotopes fell in the 0 to 25 per cent class interval*; cesium, strontium, barium-lanthanum, 25 to 75 per cent; cadmium and tungsten as tungstate, 75 to 90 per cent; cerium-praseodymium, 90 to 95 per cent; and scandium, yttrium, zirconium-niobium, phosphorus as phosphate, and chromium, 95 to 99 per cent.

---

* The values reported do not indicate the minimum and maximum range of removals but only that the values obtained fell within the interval indicated. The intervals that have been selected are 0 to 25, 25 to 75, 75 to 90, 90 to 95, 95 to 99, and 99+ per cent.

b. Filtration. - Filtration through sand was generally ineffective, except in the case of colloidal materials such as scandium, yttrium, and zirconium-niobium.

c. Ion exchange materials. - Greensand-tungsten as tungstate, 0 to 25 per cent; yttrium, 25 to 75 per cent; barium-lanthanum and scandium, 95 to 99 per cent; and strontium, 99+ per cent. Cation exchange resins (IR-120 on either the H^+ or Na^+ cycles) - tungsten as tungstate, 0 to 25 per cent; zirconium-niobium, 25 to 75 per cent; yttrium, 75 to 90 per cent; cadmium, barium-lanthanum, and scandium, 95 to 99 per cent; and cesium and strontium, 99+ per cent. Anion exchange resins (Dowex-1 on either the OH^- of Cl^- cycles) - cesium, strontium, and cadmium, 0 to 25 per cent; barium-lanthanum, 25 to 75 per cent; scandium, yttrium, and tungsten as tungstate, 95 to 99 per cent; and zirconium-niobium and iodine as iodide, 99+ per cent. Mixed ion exchange resins (IR-120 on either the H^+ or Na^+ cycles plus Dowex-1 on either the OH^- or Cl^- cycles) - scandium, yttrium, and tungsten as tungstate, 95 to 99 per cent; and cesium, strontium, cadmium, barium-lanthanum, zirconium-niobium, phosphorus as phosphate, iodine as iodide, and cerium-praseodymium, 99+ per cent.

d. Lime-soda ash softening (dosages of 10 gpg lime and 10 gpg soda ash). - Tungsten as tungstate, 0 to 25 per cent; zirconium-niobium, 75 to 90 per cent; strontium, barium-lanthanum, scandium, and yttrium, 90 to 95 per cent; and cadmium, 99+ per cent.

e. Phosphate coagulation (100 ppm PO_4). - Tungsten as tungstate, 0 to 25 per cent; antimony, 25 to 75 per cent; strontium, 75 to 90 per cent; and zinc, scandium, yttrium, zirconium-niobium, and cerium-praseodymium, 99+ per cent.

f. Metallic dusts (1000 ppm Fe dust slurried 90 minutes). - Cesium, 0 to 25 per cent; iodine as iodide, 25 to 75 per cent; barium-lanthanum and cobalt, 90 to 95 per cent; yttrium and ruthenium, 95 to 99 per cent; and zirconium-niobium, phosphorus as phosphate, and cerium-praseodymium, 99+ per cent.

From these data it will be seen that specific radioisotopes vary considerably in their amenability to removal by the process indicated. In the attached table some calculations have been made to show the amount of treatment and the process to be employed to bring about this degree of treatment for various MPC values, i.e., emergency values for 1 day and 30 day exposure, as well as long time values. These calculations have been made under the assumption that the initial activity present is 1 μc/cc.

Degree of Treatment Required to Meet Emergency or Long-Time MPC Values for Specific Radioisotopes Assuming 1 uc/cc Initially Present

Radioisotope	MPC Values-uc/cc Emergency 1 day	30 day	Long Time	Per Cent Removal Required MPC_1	MPC_{30}	MPC_{LT}	Degree of Treatment Required to Meet MPC Value MPC_1	MPC_{30}	MPC_{LT}
Cs^{137}	$5x10^{-1}$	$2x10^{-2}$	$1.5x10^{-3}$	50	98	99.85	CL,CX	CX	MX
Zn^{65}	$9x10^{-1}$	$3x10^{-2}$	$6x10^{-2}$	11	97	94	C	PC	PC
Sr^{90}-Y^{90}	$1x10^{-3}$	$5x10^{-5}$	$8x10^{-7}$	99.9	99.995	99.99992	IX	D(?)	D(?)
Ba^{140}-La^{140}	$2x10^{-1}$	$1x10^{-2}$	$2x10^{-3}$	80	99	99.8	SO,CX	MX	MX
Sc^{46}	$2x10^{-1}$	$8x10^{-3}$	$3x10^{-1}$	80	99.2	70	C	PC	C
Y^{90}	$5x10^{-1}$	$1x10^{-1}$	$2x10^{-1}$[a]	50	90	80	C	MX,PC	C
Zr^{95}-Nb^{95}	$3x10^{-1}$	$1x10^{-2}$	$4x10^{-3}$[b]	70	99	99.6	C	PC	PC,MX
P^{32}	$5x10^{-1}$	$3x10^{-2}$	$2x10^{-4}$	50	97	99.98	C	C,MX	MX
Sb^{124}	$2x10^{-1}$	$8x10^{-3}$	--	80	99.2	--	MX(?)	MX(?)	--
Cr^{51}	$3x10^{1}$	$1x10^{0}$	$5x10^{-1}$	0	0	50	None	None	C
I^{131}	$9x10^{-3}$	$8x10^{-4}$	$3x10^{-5}$	99.1	99.92	99.997	AX	Da	Da(?),MX
Co^{60}	$2x10^{-1}$	$6x10^{-3}$	$2x10^{-2}$	80	99.4	98	CL,MD	MX(?)	MX(?)
Ru^{106}-Rh^{106}	$1x10^{-3}$	$2x10^{-5}$	$1x10^{-1}$	99.9	99.998	90	MX(?)	Da	MX(?),MD
Ce^{144}-Pr^{144}	$2x10^{-1}$	$7x10^{-3}$	$4x10^{-2}$	80	99.3	96	C	PC,MX	PC

---

[a] Y^{91} MPC value, [b] Nb^{95} MPC value, CX-cation exchange, AX-anion exchange, MX-mixed bed ion exchange, IX-ion exchange, CL-clay addition, C-coagulation, settling, and filtration, PC-phosphate coagulation, SO-softening, MD-metallic dusts, D-distillation, Da- Distillation in alkaline medium, (?)-stated treatment may be effective, no data available, None-no treatment required, 1day,30day-1 day and 30 day exposure, resp., MPC_1,MPC_{30}, MPC_{LT}-emergency MPC, 1 day exposure, emergency MPC, 30 day exposure, and long time MPC, respectively.

Field Decontamination

In the small-scale decontamination of water supplies, mixed bed ion exchange resin columns would be of positive value in giving removals in the order of 99.9 per cent or more. If practicable, passage of the contaminated water through soils (silts and clays, humus, etc.) may result in substantial, if not complete, removals of the radiocontaminants. One of the most promising small-scale units contained beds of steel wool, burnt clay (clay pellets), activated carbon, and cation-anion exchange resins in series. A water containing a mixture of radioactive materials was passed through this unit and a removal of 99.996 per cent was obtained for a volume of 30 liters of water.

Distillation units should be capable of effecting removals of the non-volatile radioactive materials of 99.99 to 99.99999 per cent which represents decontamination factors* of 10^4 to 10^7.

Instrumentation

With respect to the Board's recommendations pertaining to instrumentation, two units have been developed for the continuous monitoring of low levels of radioactivity in liquids. The first consists of a G-M tube surrounded by a bed of cation exchange resin. This bed of resin concentrates the radioactive materials and the build-up in activity is recorded. It has been demonstrated that this instrument is capable of measuring activities due to cationic radioisotopes in the order of 10^{-6} to 10^{-7} μc/cc for rain water samples.

---

* The decontamination factor (D.F.) is calculated from the ratio of influent activity to the effluent activity.

In the second type of instrument, the contaminated water flows under centrifugal force along the wall of a confining cylinder and a specially prepared G-M tube is lowered into the air space. With this instrument concentrations of beta activities of about 10^{-6} μc/cc and alpha activities of about 4×10^{-5} μc/cc have been measured.

Several commercial continuous monitors are available capable of reading activities in the order of 10^{-2} μc/cc which is below the 9×10^{-2} μc/cc emergency value for 10-day use immediately after the blast.

In the evaluation of commercially available monitors for use by waterworks personnel in time of emergency, G-M survey meters and electroscope instruments have been utilized. Commercial G-M survey meters could be used for monitoring liquids of activities in the range of 10^{-1} to 10^{-4} μc/cc provided that the survey meters were calibrated with samples of similar radiochemical composition and under the same conditions of geometry. Landsverk electroscopes were found sensitive in the range 10^{-2} to 10^{-6} μc/cc for alpha and 1 to 10^{-4} μc/cc for beta emitters. The response of this instrument was found to vary with the type and energy of the radiation. Comparison of the two types of instruments for emergency use indicates that both types are suitable for measuring activity concentrations in the emergency tolerance range. On the basis of cost and availability, either instrument should be satisfactory. The electroscope offers some advantage in that is is sensitive to alpha radiation as well as beta radiation and also it is easy to maintain. On the other hand, the G-M survey meter is a more versatile instrument and would be useful for other monitoring needs during an emergency.

TABLE OF CONTENTS

Section	Page No.
I. Abstract	I- 1
II. Research on the Decontamination of Radioactive Waters	II- 1
Development of Research Program	II- 1
Present Status of the Program	II- 6
III. The Problem of Radioactivity in Watercourses	III- 1
Contamination from Nuclear Bombs	III- 2
Disposal of Wastes from Atomic Energy Installations	III- 4
Disposal of Wastes by Off-Commission Users	III- 6
Maximum Permissible Concentration of Radioisotopes in Water	III- 7
References	III-15
IV. Concentration of Radioactivity by Natural Agents	IV- 1
Physical Factors	IV- 1
Chemical Factors	IV- 3
Biological Factors	IV- 5
References	IV-11
V. Water Decontamination	V- 1
Selection of Radioelements to be Investigated	V- 2
Laboratory Studies	V- 5
Procedure	V- 5
Addition of Clay Particles (Turbidity)	V- 6
Coagulation and Settling	V-12
Sand Filtration	V-21
Lime-Soda Softening	V-22
Barium, Strontium, and Cadmium (Group II Elements)	V-24
Yttrium and Scandium (Group III Elements)	V-27
Zirconium-Niobium (Group IV and V)	V-27
Cesium (Group I) and Tungsten (Group VI)	V-27
Ion Exchange	V-28
Two-bed Deionization	V-29
Monobed Deionization	V-29
Cation Exchange	V-29
Phosphate Coagulation	V-36
Metallic Dusts	V-40

V. Water Decontamination (continued)

Pilot Plant Studies V-42
  Coagulation, Sedimentation, and Filtration V-42
  Softening V-48
Emergency-Type Water Treatment Units V-50
  Filtr-Ion Unit V-52
  Multi-Bed Decontamination Unit V-53
  Field Emergency Units V-55
  Field Distillation Units V-56
Summary of Water Decontamination Results V-56
References V-60

VI. Instrumentation VI- 1
Column Type Monitor VI- 2
Centrifugal Type Monitor VI- 3
Other Types of Continuous Monitors VI- 5
Monitoring Systems VI- 7
  Waste Monitoring System at ORNL VI- 7
  Multiple Point Monitoring System VI- 9
River and Lake Survey Equipment VI-10
Well-Logging Equipment VI-12
Miscellaneous Measurements of Radioactivity VI-13
Emergency Water Monitoring VI-14
Summary VI-16
References VI-18

Appendix A - Staff Members - Radioactive Liquid Waste Research and Development Section

Appendix B - Oak Ridge Tap Water Analyses

LIST OF TABLES AND FIGURES

Table No. or Figure No. — Page No.

III- 1. Maximum Permissible Concentrations of Radioisotopes in Water III- 8

IV- 1. Sources and Magnitude of Background Radiations IV- 2
2. Concentration of Radioactive Materials by Algae IV- 7
3. Concentration of Radioactive Materials by Algae IV- 8
4. Takeup of Activity (Sr^{89}) from Water by Live and Dead Fish IV-10

V- 1. Radioelements Suitable for Study V- 4
2. Effect of Turbidity on Removal of Radioisotopes V- 8
3. Coagulation and Settling Results - Jar Test Studies V-14
4. Ferric Chloride-Limestone Coagulation Results V-18
5. Alum and Limestone Coagulation Results V-19
6. Laboratory Sand Filtration Results V-23
7. Approximate Minimum Combined Dosage of Lime and Soda Ash to Give Stated Removal V-25
8. Percentage Removal Results of Individual Isotopes with Ion Exchange Materials V-31
9. Removal of Radioactive Contaminants from Water by Combined Anion-Cation Resin V-35
10. Removal of Radioisotopes by Phosphate Coagulation V-38
11. Removal of Radioactive Contaminants by Metallic Dusts V-41
12. Removal of Radioisotopes - Conventional Water Treatment Processes V-45
13. Removal of Mixed Fission Products - Conventional Water Treatment Processes V-46
14. Removal of P^{31} and P^{32} by a Rapid Sand Filtration Pilot Plant V-47
15. Removal of Fission Products Activity by Coagulation, Settling, and Filtration V-49
16. Pilot Plant and Jar Test Results - Softening Experiments V-51
17. Removal of Radioactive Contaminants - Lauderdale and Emmons Unit V-54
18. Optimum Removal of Radioisotopes from Water V-58

Figure No.

V- 1. Flow Diagram of ORNL Water Treatment Pilot Plant V-43

SECTION I

ABSTRACT

This interim progress report incorporates the latest information on water decontamination and related problems. The development of the research program at ORNL, the physical plant, and the present status of the program are indicated. Consideration is given to probable sources of radioactive contamination along with their potential health hazard should the contamination find its way into drinking water supplies. Among the sources discussed are the nuclear bomb, the atomic energy installations, and the off-commission users of radioisotopes.

Experimental results are included which indicate the effects of natural agents (physical, chemical, and biological) which concentrate radioactive materials and thus remove them from the waste streams. The results of water decontamination studies are grouped under three major headings - laboratory studies, pilot plant studies, and emergency water treatment units. These findings are discussed according to the treatment process involved.

The degree of removal of radio-contaminants from liquids by water treatment plant processes is contingent upon a wide variety of factors, which are associated with the chemical form of the contaminant as well as the general nature of the liquid medium. With regard to the decontamination of water under emergency conditions utilizing normal water treatment processes or feasible modifications

thereof, some removal of radioactive material will be effected. However, such removal may or may not be adequate and should be evaluated on the basis of the MPC value for the individual radioisotopes or on the basis of the weighted MPC value for the mixture under consideration, in order to determine whether a health hazard exists. For the removal of individual radioisotopes from waters, modified treatment methods are set forth which might be applied to the treatment of the radioactive materials.

The final section of the report describes the activities of the ORNL group in the evaluation and adaptation of commercially available instruments along with discussions relating to the development of new instruments to meet specific needs. Some consideration is given to the use of commercial instruments for measuring radioactive concentrations in water under emergency conditions.

SECTION II

RESEARCH OF THE DECONTAMINATION OF RADIOACTIVE WATERS

This review is intended as an interim progress report on the cooperative research investigations of the Radioactive Liquid Waste Research and Development Section (RLWRDS), Health Physics Division, Oak Ridge National Laboratory (ORNL), related to the decontamination of radioactive water supplies, and includes information available in the unclassified literature. It presents, in addition, a brief description of the evolution of the radioactive liquid waste disposal program and describes the physical plant and facilities available at ORNL for studies on this, and related subjects.

Development of the Research Program

On July 1, 1948, the Radioactive Liquid Waste Research and Development Section (RLWRDS) was organized within the framework of the Health Physics Division, ORNL, with Mr. Roy J. Morton as Section Leader. The primary objectives of this Section were to evaluate existing water treatment processes for the decontamination of drinking water rendered unsafe by contamination with radioactive substances, to consider new processes, and to develop methods for the safe and satisfactory disposal of radioactive wastes into the environment.

The program, sponsored by the Atomic Energy Commission, (AEC), was carried on cooperatively with other interested agencies and included participation by the Environmental Health Center, U. S.

Public Health Service; the Health and Safety Division, Tennessee Valley Authority (TVA); the Corps of Engineers, Department of the Army; the U. S. Geological Survey; the U. S. Weather Bureau, and the University of Tennessee.

In an exchange of correspondence between the Surgeon General, Public Health Service, and the General Manager, Atomic Energy Commission, Oak Ridge Operations, in December, 1949, and January, 1950, it was agreed that the Public Health Service would undertake technical sponsorship of an intensified cooperative research program on the decontamination of radioactive water supplies to be carried out at the Oak Ridge National Laboratory.

In 1950, the Surgeon General, Public Health Service, established the Technical Advisory Board for Research on the Decontamination of Radioactive Waters, to provide broad guide lines for the research program. The Advisory Board includes representatives from the Atomic Energy Commission, the Public Health Service, the Department of Defense, the American Water Works Association, and the Conference of State Sanitary Engineers. At its first meeting on October 6, 1950, the Board recommended that research priority be given to the evaluation of the effectiveness of (1) military type rapid coagulation and filtration apparatus in removing mixed fission products*, (2) standard water treatment processes and variations thereof in the removal of the mixed fission products and individual radioisotopes; and as a supplement to this, evaluation of processes suitable for small scale field decontamination of water supplies. The Board further recommended that concurrent attention be given to the problems of developing (1) suitable instruments for continuous monitoring of water supplies and the detection of low level radioactive contamination, and (2) adequate methods and standards for the detection of radioactive contaminants in water under emergency conditions.

The research program has been conducted primarily in the laboratories of the Health Physics Division, Oak Ridge National Laboratory, and since late 1951, in a special research building constructed for the program. A brief historical review of the RLWEDS laboratory facilities might be of some interest. The project was conceived in preliminary form as a result of informal discussions between Dr. A. H. Holland, Director of Research and Medicine, AEC, Oak Ridge, and Dr. K. Z. Morgan, Director of Health Physics Division, ORNL, and others, in the period prior to July, 1948. At a meeting in August, 1948, the building project was discussed and it was decided that plans and cost estimates be prepared. As a result, the RLWED Section outlined types of facilities and equipment required. The problems involved were clearly delineated, and a proposed research program for attacking them on a broad and fundamental basis was described in a report entitled "Proposal for Evaluating the Impact of Waterborne Radioactive Contaminants on Sanitary Engineering Facilities" (ORNL 48-10-377, October 29, 1948).

---

* Since the results of these studies are contained in classified reports prepared by the Engineer Research and Development Laboratory, this material is not included as a part of the "Interim Report". For those interested the material may be found in the following reports:

1 Lacy, William J., and Lindsten, Don C., "Purification of Water Contaminated with Radioactive Materials". Report on Project 8-75-05-008 Submitted to Chief of Engineers, U. S. Army. (1 July, 1952).

2 Lacy, William J., and Lindsten, Don C., "Removal of Radioactive Materials from Contaminated Water by Thermocompression Distillation". Interim Report on Project 8-75-05-008 Submitted to Chief of Engineers, U. S. Army. (31 December 1952).

3. Lacy, William J., and Lindsten, Don C., "Ion Exchange Materials for Radioactive Water Decontamination". Interim Report on Project 8-75-05-008 Submitted to Chief of Engineers, U. S. Army. (1 May 1953).

By the end of 1949, detailed equipment and building cost estimates had been prepared. The cost of the project was borne jointly by the AEC, National Military Establishment, USPHS, and ORNL. On January 12, 1951, ground was broken for the new facility. The building was completed ten months later, at which time the RLWEDS Sanitary Engineering Processes Group and certain other Section Personnel and equipment were transferred to it. Other personnel in the Section are still located in the main Health Physics building.

The new research building contains a laboratory, pilot plant area, storage space, and office facilities. The laboratory room on the ground floor has a floor area of approximately 1200 square feet and was designed to facilitate radiochemical and sanitary analyses. In addition to the usual laboratory type benches and hoods, two constant temperature rooms were provided which may be used for either refrigeration or incubation in controlled biological investigations. The pilot plant section has a floor area of approximately 1800 square feet and a maximum ceiling height of 23 feet. Located in this area at present are: a water treatment pilot plant, a water softening unit, small scale trickling filters, jar test mixing devices, small columns for filtration and ion exchange studies, counting equipment, and other related apparatus. Three storage tanks, each of approximately 5000 gallons capacity, are located outside the building for preparation of various types of raw water for process use. Office facilities are located on the mezzanine floor above the laboratory area.

At present, the new building is staffed with three sanitary engineers and two chemists from the USPHS, and a chemist and physical science aide of the Health Physics Division, ORNL. Other staff members included in the RLWRDS located in the Health Physics building are: a sanitary engineer, chemist, biologist, physicist, physical science aide, and a secretary (all members of the Health Physics

Division, ORNL); a scientist and a physicist of the USPHS; and a chemist and a physical science aide of the Corps of Engineers, U. S. Army*.

Present Status of the Program

There are several basic categories into which the activities of the HLWRDS can be placed. One of these, the problem of water decontamination, has thus far received the most attention and those phases pertaining to the evaluation of the efficacy of conventional water treatment processes in the removal of single radioisotopes are virtually complete. Work will continue on such phases of the problem as the cooperative military project, the development and evaluation of emergency-type units, and non-conventional methods of treatment. A re-examination of the more effective methods of treatment and confirmatory work on laboratory and pilot plant results is contemplated.

Another important phase of the over-all program is the treatment of radioactive wastes. The application of suitable treatment processes at the source would minimize radioactive contamination of the environment, simplify treatment procedures, decrease the size of treatment units required, and in all probability, reduce the cost of treatment. Sanitary engineering processes may be adapted to the treatment of low-level radioactive wastes because their major applications have been in the treatment of large volumes of liquids containing small concentrations of impurities. The high volume - low activity process wastes that would require treatment probably fall into this category.

---

* For a complete listing of personnel who are or who have been associated with the project see Appendix "A".

Studies are also in progress to evaluage the effect of natural agents such as clays, soils, and biological agents upon removal of radioactive material. This is a significant problem because low-level radioactive wastes are now discharged into streams or stored in natural lagoons near the operational areas. Methods of treating the radioactive wastes presently discharged by ORNL into the White Oak Lake system are being studied. Towards this end, a collection and analysis of waste data regarding flow patterns, radioactivity concentrations, and chemical and radiochemical composition are being made. In addition, sewage treatment processes, sludge disposal problems, and the evaluation of combined processes for small users have been investigated to some extent.

Certain groups in RLWRDS devote their efforts to an improvement of instrumentation techniques. There continues to be a need for the critical evaluation of commercially available radiation detectors for emergency use, and developments are being made for the continuous monitoring of waste streams. Specialized instrumentation for the detection of very low levels of activity has been studied. These developments are especially important when information on "before-and-after" type field surveys is required.

The application of the maximum permissible concentration (MPC) concept to an evaluation of the environmental hazards of radioactive waste disposal is being emphasized. Cognizance must be taken of the relative hazards associated with single isotopes or mixtures thereof and, particularly, the ease or difficulty encountered in their removal from waters.

Of necessity, the Section must take an active interest in such fields as the use of radioactive tracers as research tools in sanitary engineering and other related studies, in the disposal of non-radioactive wastes peculiar to the atomic energy industry, in the training of sanitary engineering personnel in this field, and in the ultimate disposal of radioactive waste materials. In addition, it should disseminate information pertaining to its fields of interest.

SECTION III

THE PROBLEM OF RADIOACTIVITY IN WATERCOURSES

There are several sources from which radioisotopes might find their way into watercourses and subsequently be encountered in drinking water supplies. The source of such contamination will depend primarily on whether peacetime or wartime uses of radioactive materials are considered. The primary sources of radio-contaminants other than those occurring naturally, are as follows: (1) use of nuclear bombs; (2) radiological warfare agents (classified and will not be discussed herein); (3) disposal of wastes from Atomic Energy Commission installations; (4) disposal of wastes by Off-Commission (non-AEC) users, where radioisotopes are employed in research, medicine, or industry.

The latter two sources, although generally considered to have major importance during peacetime, could contribute far more damaging release of radioactive materials from the health hazard standpoint than the first two sources in the event of bombing or destruction of radioisotope storage facilities, particularly storage lagoons for liquid wastes.

The major pertinent radio-contaminants (contributed by each of these sources) are discussed. Following this discussion, a general approach to the problem of radioactivity in water based on the maximum permissible concentrations of radioisotopes in water is given.

Contamination from Nuclear Bombs

Morgan and Straub(1) have stated that an underwater or underground explosion of a nuclear bomb or heavy fall-out due to unusual meterological conditions following an air burst are the only instances where a bomb explosion would result in appreciable surface contamination. They have also indicated that nearly all (95% or more) of the residual activity would be due to fission products.

On the basis of the fission product decay rates and maximum permissible concentrations which have been established for radioisotopes, it is possible to estimate the relative health hazard of specific radioelements at various time intervals following detonation of a nuclear bomb. An estimate as to which radioelements present the principal health hazard has been determined for time intervals of one day, one week, one month, one year, and ten years following a bomb explosion(2). These radioelements would require relatively greater removals (percentage wise to meet the lower MPC values of these materials) in the event significant contamination of a water supply occurred following a nuclear explosion. The more critical elements involved for time intervals up to one month include the following (in descending order of importance): tellerium, strontium, iodine, barium, niobium, xenon, cerium, rhodium, praseodymium, and yttrium. For the one year and ten year periods, strontium is by far the most critical element followed by niobium. From the standpoint of contamination of water supplies by nuclear bombing, most concerted effort should be devoted to devising treatment procedures which might effect substantial removals of these elements. An inherent difficulty in developing such treatment procedures is the fact that the chemical forms of these various radioelements in a bomb blast mixture have not been clearly defined. It is believed that oxides of the metals are formed and some calculations have been made based on these observations.

The chemical form or valence state of particular radioelements, as for example, ruthenium, chromium, etc., is of prime importance in determining removal efficiencies by treatment processes. In some analytical studies with chromium (not the radioisotope), Seal(3) showed that trivalent chromium was readily removed by coagulation with alum, ferric chloride, and ferrous sulfate, whereas in the case of hexavalent chromium practically no removal of the chromium was obtained following coagulation with alum and ferric chloride. Complete removal of the hexavalent form was obtained following coagulation with ferrous sulfate according to the following equations:

$$Fe^{++} + Cr_2O_7^{=} + 14\ H^{+} \longrightarrow Fe^{+++} + 2\ Cr^{+++} + 7\ H_2O,$$

and

$$Cr^{+++} + 3\ (OH)^{-} \longrightarrow Cr\ (OH)_3 \downarrow$$

It is presumed that similar reactions will take place when chromium radioisotopes are studied. Brockett(4) has shown that $Ru^{106}Cl_2$ as received from the Operations Division, ORNL, is almost completely removed following passage through a clay soil, yet the Ru^{106} contained in a lagooned waste material when passed through the same soil column is not removed. It is believed that the chemical form of the ruthenium in the latter case is different from the $Ru^{106}Cl_2$. This is quite possible in view of the heterogeneous nature of the lagooned waste materials.

Disposal of Wastes from Atomic Energy Installations

Under continued present peacetime conditions, it is unlikely that any appreciable health hazard will result from the discharge of waste radioactive materials by AEC controlled installations. The reactor program, however, may be accelerated, due to wartime emergency or private power developments, and significant quantities of wastes might be discharged some time in the future. It is probable that only those wastes of relatively high volume-low activity need be discharged directly since additional storage space (together with further concentration) could likely be provided for the more concentrated wastes of low volume. The cooling waters used for reactor cooling, such as at the Hanford Works, constitute the largest volume of wastes. The radioelements to be considered in such instances include sodium, phosphorus, iron, calcium, and sulfur. By treating the water prior to use, it is possible to substantially reduce the amount of radioactivity induced as the water passes through the reactor.

The storage of waste liquids containing radioisotopes also may give rise to water supply problems. This practice is common in atomic energy installations to permit decay of activity prior to disposal. Leakage of such materials into ground waters may occur. Storage facilities may be damaged in time of war or other catastrophe and this might result in the discharge of slugs of waste to watercourses. As the number of reactors being constructed throughout the country increases and storage facilities for waste materials become more widespread, the possibilities of inadvertent discharge of such waste becomes greater.

In general, these wastes consist of comparatively long-lived isotopes in much the same proportion as in the fission product mixture from a bomb blast. Studies are currently under way to determine which radioisotopes might likely leach through soil barriers. Upon completion of these studies, it should be possible to evaluate the potential hazard from such contamination.

Disposal of Wastes by Off-Commission Users

The prospective hazard due to widespread usage of radioisotopes by non-AEC installations is governed by several factors. Foremost is the fact that radioisotopes are not distributed to such installations until the AEC is satisfied that the user is thoroughly familiar with the required safeguards to prevent the material from becoming a hazard. This applies to the disposal of any radioactive materials as well. The quantity (curies) of isotopes being utilized to date is relatively small when the amount of dilution water avilable is considered. Should the quantities of isotopes used at Off-Commission sites increase significantly, it would be possible to forecast which isotopes might become critical when disposed into watercourses. A method for forecast has been outlined by Straub(5), who has further indicated that for shipments up to December, 1950 strontium was by far the most critical radioisotope involved on the basis of dilution volume required to satisfy the maximum permissible concentration (MPC). Such evaluation should be made periodically, both overall and for pertinent local areas, so that suitable waste treatment and disposal methods can be devised to reduce the hazard. In this way, radioisotopes may be used without any significant health hazard resulting from the discharge of waste materials.

To summarize, the problem of radioactivity in watercourses is dependent upon such unpredictable factors as the existence of war- or peacetime conditions in this country and the rate and manner of expansion of work in the atomic energy field. Therefore, it is necessary to continually search out specific instances where radioactive contamination of watercourses exists or may become significant.

Maximum Permissible Concentrations of Radioisotopes in Water

The Subcommittee on Internal Dose of the National Radiation Protection Committee under the chairmanship of Dr. K. Z. Morgan, Director, Health Physics Division, ORNL, has prepared and published considerable information on the maximum permissible concentrations (MPC) of radioisotopes under peacetime (long term) exposure(6). Only the values reported for the various concentrations of radioisotopes in water are of interest in this discussion. These, along with more recent values for other radioisotopes in water(7), are given in Table III - 1. General values of MPC for mixtures of radioisotopes have been selected by this same committee(6), and are also listed in Table III - 1. These values may be used for short periods of exposure (for a few months) regardless of the radioisotope and are believed safe for extended use of all radioisotopes listed in Table III - 1 with the possible exception of Ra^{226}. In operations where the level of radioactive contamination in water never exceeds the general values given (unidentified radioisotopes), it may be possible to rely on simple gross contamination

TABLE III - 1

Maximum Permissible Concentrations of Radioisotopes in Water(6)(7)(11)

Radioisotope	MPC Values-uc/cc Exposure Times 1 day	 30 day	 Life
Unidentified β,γ	--	--	1x10^{-7}
Unidentified α	--	--	1x10^{-7}
H^{3} (HTO,H$_2^{3}$O)	5x10^{1}	2x10^{0}	2x10^{-1}
Be^{7}	8x10^{0}	3x10^{-1}	1x10^{0}
C^{14} (CO$_2$)	1.5	5x10^{-2}	3x10^{-3}
F^{18}	1x10^{2}	1x10^{2}	9x10^{-1}
Na^{24}	5x10^{-1}	3x10^{-1}	8x10^{-3}
P^{32}	5x10^{-1}	3x10^{-2}	2x10^{-4}
S^{35}	3.6	1x10^{-1}	5x10^{-3}
Cl^{36}	1.4	4x10^{-2}	2x10^{-3}
A^{41}			5x10^{-4}
K^{42}	1x10^{0}	7x10^{-1}	1x10^{-2}
Ca^{45}	3x10^{-1}	3x10^{-3}	5x10^{-4}
Sc^{46}	2x10^{-1}	8x10^{-3}	4x10^{-1}
V^{48}	2x10^{-2}	1x10^{-3}	5x10^{-1}
Cr^{51}	3x10^{1}	1x10^{0}	5x10^{-1}
Mn^{56}	1x10^{1}	1x10^{1}	2x10^{-1}
Fe^{55}	2	8x10^{-2}	4x10^{-3}
Fe^{59}	6x10^{-2}	3x10^{-3}	1x10^{-4}
Co^{60}	2x10^{-1}	6x10^{-3}	2x10^{-2}
Ni^{59}	4x10^{1}	1.4	2x10^{-1}
Cu^{64}	7	5	8x10^{-2}
Zn^{65}	9x10^{-1}	3x10^{-2}	6x10^{-2}
Ga^{72}	6x10^{-1}	4x10^{-1}	9
Ge^{71}	3x10^{1}	2	9
As^{76}	5x10^{-1}	2x10^{-1}	2x10^{-1}
Rb^{86}	4x10^{-1}	2x10^{-2}	3x10^{-3}
Sr^{89}	1x10^{-1}	5x10^{-3}	7x10^{-5}
Sr^{90}-Y^{90}	1x10^{-3}	5x10^{-5}	8x10^{-7}
Y^{91}	5x10^{-1}	2x10^{-2}	2x10^{-1}
Nb^{95}	6x10^{-1}	2x10^{-2}	4x10^{-3}
Mo^{99}	8x10^{-1}	2x10^{-1}	14
Tc^{96}	2x10^{-1}	4x10^{-2}	3x10^{-2}
Ru^{106}-Rh^{106}	1x10^{-3}	2x10^{-5}	1x10^{-1}
Rh^{105}	1	5x10^{-1}	1.5x10^{-2}
Pd^{103}-Rh^{103}			1x10^{-2}
Ag^{105}			2
Ag^{111}			4
Cd^{109}-Ag^{109}			7x10^{-2}
Sn^{113}	4x10^{-1}	1x10^{-1}	2x10^{-1}
Te^{127}			3x10^{-2}
Te^{129}	4x10^{-1}	2x10^{-2}	1x10^{-2}
I^{131}	9x10^{-3}	8x10^{-4}	3x10^{-5}
Xe^{133}	2	3x10^{-1}	4x10^{-3}
Xe^{135}	3	3	1x10^{-3}
Cs^{137}-Ba^{137}	5x10^{-1}	2x10^{-2}	1.5x10^{-3}
Ba^{140}-La^{140}	2x10^{-1}	1x10^{-2}	2x10^{-3}
La^{140}	4x10^{-1}	1x10^{-1}	1
Ce^{144}-Pr^{144}	2x10^{-1}	7x10^{-3}	4x10^{-2}
Pr^{143}	1	6x10^{-2}	4x10^{-1}
Pm^{147}	4	1x10^{-1}	1
Sm^{151}	1x10^{1}	5x10^{-1}	2x10^{-1}
Eu^{154}			3x10^{-2}
Ho^{166}			23
Tm^{170}			2x10^{-1}
Lu^{177}			24
*Ta^{182}			1x10^{-1}
*W^{181}			2x10^{-1}
Re^{183}			2x10^{-1}
Ir^{190}			1x10^{-2}
Ir^{192}			9x10^{-4}
*Pt^{191}			6x10^{-3}
*Pt^{193}			5x10^{-3}
*Au^{196}			5x10^{-3}
Au^{198}			3x10^{-3}
Au^{199}			7x10^{-3}
*Tl^{200}			2x10^{-2}
*Tl^{201}			7x10^{-2}
*Tl^{202}			2x10^{-2}
*Tl^{204}			7x10^{-3}
Pb^{203}			1x10^{-1}
Po^{210}(sol.)			3x10^{-5}
At^{211}			2x10^{-6}
Rn^{222}+dr.			2x10^{-6}
Ra^{226}+1/2 dr.			4x10^{-8}
Th^{234}			3
U-nat.(sol.)			7x10^{-5}**
U^{233}(sol.)			1.5x10^{-4}
Pu^{239}(sol.)	3x10^{-2}	2x10^{-3}	1.5x10^{-6}
Am^{241}			1x10^{-4}
Cm^{242}			9x10^{-4}

---

* New values which have not yet been submitted to the National Committee on Radiation Protection.

** Value based on chemical toxicity rather than on radiation hazard.

measurements rather than performing radiochemical analyses which are both expensive and time consuming. For permanent installations that dispose of large quantities of radioactive waste, the MPC values in Table III - 1 should probably be reduced by a factor of 10, as suggested in the report of the National Radiation Protection Committee(6).

The Atomic Energy Commission(8) recently released tentative limits of 9×10^{-2} uc/cc for beta and gamma radioactivity, and of 5×10^{-3} uc/cc for alpha activity for the consumption of drinking water for 10 days use immediately following a nuclear disaster. If the contaminated water is to be consumed over a period of thirty days values one-third as high as those above have been suggested. Similar values have been reported by Marsh(9). Values somewhat lower than these have been suggested by Bale(10), namely: 2.5×10^{-1} uc/cc for beta activity and 3.7×10^{-2} uc/cc for alpha activity for water to be consumed for ten days during the first 50 days following a bomb blast. After some study of the problem, Morgan and Straub(1) found that a relatively simple equation could be developed which would give reasonable and safe values of MPC in water for internal exposure at any time following an atomic explosion. The equation, which is of the form

$$MPC = 10^{-3}\ t^{-1.2}$$

where MPC is in uc/cc and t is in days after the blast, was designed to be applicable within the limits of about 0.3 days to about 3×10^3 days. More recently Morgan and Straub(11) have computed emergency MPC values for a number of radioisotopes. These values are reported in Table III - 1. In addition they have indicated equations and methods which will permit calculation of the MPC values of mixtures of radioisotopes.

The MPC concept is an important one and is a necessary adjunct to any consideration of removal efficiency. As such it should be thoroughly understood by the sanitary engineer and water works official. The following illustrative examples bring out four points which are very important: (1) the initial concentration of the mixture must be known, (2) knowledge of the composition of the main constituents is necessary for a true evaluation of the hazard of the waste, in other words, a radiochemical analysis of the waste is required, (3) the MPC value must be known for the given radioisotopes in question to evaluate the true removal or efficiency of the process or processes under investigation, and (4) the remaining or residual activity must be known, since this determines whether or not the treatment process actually met the concentrations delineated by the MPC analysis.

Assume an unknown mixture of radioisotopes with a gross beta concentration of 10^{-3} uc/cc. This level of activity is 10^{4} times higher than is permissible for an unknown mixture of radioisotopes (MPC value - 10^{-7} uc/cc). If a water containing this level of activity was to be treated to the 10^{-7} uc/cc level, it would be necessary to provide for a removal of 99.99 per cent. Such removal could not be obtained by the conventional water treatment processes, and it would be necessary to resort to more costly procedures such as ion exchange or distillation.

Suppose a radiochemical analysis of this waste material was made and it was found to contain the following radioelements: Y^{91}, 20 per cent; Ru^{106} -Rh^{106}, 20 per cent; Sn^{113}, 20 per cent; La^{140}, 20 per cent; and Pm^{147}, 20 per cent. The equivalent or weighted MPC value is found to be 2.2 x 10^{-1} uc/cc, which is considerably higher than the givin initial concentration of 10^{-3} uc/cc, and certainly there would be no radiological hazard from the consumption of this water as is. Now modifying the assumed composition by substituting 20 per cent Sr^{90} -Y^{90} for the Sn^{113}, the equivalent MPC value for this combination of radioisotopes is found to be 4 x 10^{-6} uc/cc, which is considerably lower than the 10^{-3} uc/cc initially present in the waste. Obviously, to avoid a possible health hazard, it would be necessary to treat this waste before use as a source of drinking water.

Percentage removal values per se have little significance as will be borne out by a further comparison of these same illustrative examples. Here again the value of the MPC concept will be shown for assessing the real hazard from a mixture of radioisotopes. Taking the two mixtures indicated above, a set of removal values for each of the constituents in the two mixtures is assumed as follows: Y^{91}, 98.0 per cent; Ru^{10} -Rh^{106}, 99.0 per cent; Sn^{113}, 99.0 per cent; La^{140}, 95 per cent; Pm^{147}, 99.9 per cent; and Sr^{90} -Y^{90}, 85 per cent. The weighted removal of mixture 1 is 98.18 per cent or 1.82×10^{-5} uc/cc remain in this mixture and the weighted removal of mixture 2 is 95.38 per cent or 4.62×10^{-5} uc/cc remain. Calculated MPC values for the mixtures following treatment are 3.63×10^{-1} and 1.23×10^{-6} uc/cc. With the first mixture, comparison of the final MPC value with the observed activity remaining indicates this water presents no hazard after treatment. In the case of the second mixture it will be noted that the percentage removal of activity is of the same order of magnitude as the first mixture. However, comparison of the activity remaining with the MPC value for this treated mixture indicates that the water is still hazardous for drinking purposes.

Certain practical difficulties limit the usefulness of the MPC concept in evaluating treatment efficiency. Radiochemical techniques have not been sufficiently developed to permit suitable measurement of relatively low concentrations of radioisotopes. As with macrochemical analyses, the analysis of radioactive mixtures becomes increasingly difficult as the number of radioelements increases or as the relative concentrations of elements differ greatly from one another.

Nevertheless, radiochemical analyses of liquid wastes should be made insofar as practicable. From the results obtained, the MPC value associated with the waste can be estimated. Comparison of this MPC value with the activity concentration present will indicate whether the waste is hazardous when diluted with the receiving stream or whether treatment of the waste is mandatory.

Although the MPC concept provides a realistic approach to the evaluation of water decontamination procedures, it will be noted throughout this report that removal efficiencies are expressed in the form of percentage removal or decontamination factor obtained. In studies using individual radioisotopes, comparison of the final activity concentration obtained with the MPC value for the particular radioisotope investigated is a simple matter. When mixtures of radioactive materials have been involved, generally it has been found impractical to estimate MPC values of the raw or treated waters due to the lack of radiochemical information. It should be mentioned, also, that extensive MPC data were not available until very recently, and consequently, studies performed during the early phases of the ORNL program could not be made with this approach in mind.

REFERENCES

1. Morgan, K. Z. and Straub, C. P. External and Internal Exposure to Ionizing Radiation and Maximum Permissible Concentration (MPC) of Radioactive Contamination in Air and Water Following an Atomic Explosion. AECU-2332 (April 4, 1952). Oak Ridge National Laboratory. Available from Office of Technical Services, Department of Commerce, Washington 25, D. C.

2. Pecsok, Donald A. A Review of Radioisotopes from the Standpoint of Water Decontamination for Drinking Supplies. Memorandum to the Files. (May 19, 1952).

3. Seal, Morgan S. Preliminary Studies on the Removal of Chromium by Conventional Coagulation Methods. Memorandum to the Files (October 23, 1952).

4. Brockett, T. W., Jr. Unpublished data. Oak Ridge National Laboratory, Oak Ridge, Tennessee (1953).

5. Straub, Conrad P. Disposal of Radioisotopes - Method of Evaluating Potential Hazard. Unpublished. Oak Ridge National Laboratory, Oak Ridge, Tennessee (April 14, 1951).

6. Morgan, K. Z., Chairman, Subcommittee on Internal Dose of the National Committee on Radiation Protection. "Maximum Permissible Concentration of Radioisotopes in Air, Water, and in the Human Body." NBS Handbook No. 52. National Bureau of Standards (1953). Available from the Superintendent of Documents, Washington 25, D. C.

7. Morgan, K. Z. Maximum Permissible Internal Dose. A Report of the Subcommittee on Permissible Dose for Internal Radiation of the International Commission on Radiological Protection. (To be presented at the 7th International Congress on Radiology, Copenhagen, Denmark, July 19-25, 1953).

8. U. S. Atomic Energy Commission. AEC Contract Policy and Operations. Ninth Semiannual Report to Congress. Page 19. (January, 1951). Available from Superintendent of Documents, U. S. Government Printing Office, Washington 25, D. C.

9. Marsh, J. S. Prior Planning Saves Lives in Atomic Disaster. Civil Engineering, 21: 3, 45 (March, 1951).

10. Bale, W. F. Levels of Radioactivity in Waters and Foods That Can Be Permitted Under Emergency Conditions in War Time. Unpublished. (July 24, 1950).

11. Morgan, K. Z. and Straub, C. P. Maximum Permissible Exposure Levels of Ionizing Radiation for Peacetime Operations and for Emergency Conditions. Unpublished Information. Oak Ridge National Laboratory, Oak Ridge, Tennessee (1953).

SECTION IV

CONCENTRATION OF RADIOACTIVITY BY NATURAL AGENTS

Before is is possible to estimate the net concentration of radioactive materials that has taken place as a result of physical, chemical, and biological processes, it is necessary to obtain information on the normal background radioactivity of the material in question. Both radiation and radioactive materials are naturally present in the environment, and will be found in small amounts in almost any substance that can be mentioned, including air, soil, building materials, and the human body(1). Radiation is given off by these materials and, in addition, is reaching the earth from outer space. The activity due to these sources is variable and should be determined. Cowan(1) has indicated orders of magnitude for activity from various sources, and these have been included in Table IV - 1.

Concentration or, more specifically, reconcentration, since it is assumed to take place at some time beyond the point of release, may result from certain physical, chemical, and biological processes. These will be discussed briefly.

Physical Factors

Among the physical factors active in the reconcentration of elements, sedimentation and temperature are perhaps of most concern. If radioactive materials are present in the form of discrete particulates, it is possible that combination or coalescence with other

TABLE IV - 1

Sources and Magnitude of Background Radiations(1)

Source	Magnitude uc/cc	Remarks
Cosmic radiation		About 70 per cent of total background at sea level is due to cosmic radiation.
Naturally occurring radioisotopes		Uranium and thorium series, K^{40}, Rb^{87}, Sm^{152}, Lu^{176}, Re^{187}.
Radioactivity of soils and rocks		Uranium - 6 ppm by wt. thorium - 12 ppm, radium - $2x10^{-6}$ ppm.
Radioactivity of air	10^{-10}	Radon concentration in air. Generally higher during rainfall.
Radioactivity in fresh water	10^{-9} to 10^{-10}	Varies, with values 10^{1} to 10^{3} times higher in some springs.
Radioactivity in sea water	$3.3x10^{-7}$	Due primarily to K^{40} contained in sea water

particulate matter would occur in a watercourse to form larger particles which may settle out.

Temperature may be construed as a physical factor leading to reconcentration by two opposite methods. Concentration by heating (solar evaporation) results in crystallization. The non-volatile fraction may be concentrated to any desired solids content or may be carried to a dry state. At several of the AEC installations, the method of evaporation has found extended use in controlling the volumes and concentrations of radioactive waste liquors. However, little use has actually been made of solar evaporation as a means of concentrating waste solids. Freezing may also be used as a method of crystallization or concentration in that when freezing takes place there is a tendency for impurities to be extruded, which results in concentration to a point where crystallization may occur. This phenomenon was investigated by Straub in some preliminary experiments and a concentration factor approaching 10^2 was observed in tests involving initial waste volumes of about 200 ml.

Chemical Factors

In addition to such factors as adsorption, absorption, flocculation, and precipitation, chemical factors include ion exchange, neutralization, etc. Depending upon the chemical properties of the waste solution (pH, salt content, total solids, heavy metal concentration, etc.) and the properties of the receiving stream, neutralization may occur, which results in precipitation of heavy metal hydroxides or carbonates.

Most receiving streams contain particles of clay in the form of turbidity. Laboratory tests and field data have indicated that these clay particles, through ion exchange, adsorption, or absorption, can remove radioactive contaminants from solution. Settling of these particles will result in substantial concentration of radioactive materials. Some years ago Morgan(2) reported data estimating the activity in the mud of the White Oak Creek drainage system below ORNL. The estimated activity in mud in April 1945 was 13 per cent of the total activity that had been discharged into White Oak Creek up to that time. When a correction was made for decay of the radioisotopes present in the mud, the indication was that the mud had retained about 70 per cent of the radioactive material discharged into White Oak Creek. The amount of activity taken up by the clay is dependent upon the specific radiocontaminants present in the wastes discharged.

The phenomenon of takeup of radioactive materials by clays and soil particles has been utilized at the Hanford Works and other installations as a means of discharging waste solutions into the ground without the fear of contaminating areas at some distance from the point of discharge. Since many of the radiocontaminants attach themselves to the soil particles and are held tenaciously near their point of discharge, this offers a very satisfactory and relatively economical method of waste disposal where geological conditions are favorable.

Recently, open storage pits have been utilized at ORNL for the disposal of liquid radioactive wastes. In conjunction with this practice, laboratory studies have been made to determine the capacity of the shale-type weathered rock and soils in the vicinity of these waste pits to remove various radioisotopes. Two methods of testing were used, namely, jar tests where varying amounts of shale were mixed with solutions containing radioisotopes and column studies where these solutions were passed through small volumes of the shale. In the jar test studies(3) for a shale concentration of 40,000 ppm, removals in excess of 99 per cent were obtained for Ba^{140}, Ce^{144}, Cs^{137}, P^{32}, Ru^{106}, and Zr^{95} whereas the removal for Sr^{90} was about 77 per cent, for I^{131} about 67 per cent, and for a mixed fission products waste the removal was 98.2 per cent. Results obtained in passing two liters of radioactive solutions through columns containing 75 grams of shale were practically identical to the jar test results. However, field tests have indicated that Ru^{106} permeates comparatively large volumes of shale. This conflicting information is probably due to differences in the chemical form or valence state of the ruthenium.

Further discussion of the takeup of radioisotopes on clay particles is contained in Section V "Water Decontamination".

Biological Factors

Biological concentration is of more intrinsic interest from the standpoint of radiological health than the other factors responsible for accumulation. As reported by Warren(4) reconcentration of radioactive materials has caused some concern since it might be conceivable first to have concentration in algae, then in snails that live on the algae, then in fish that eat the snails, and finally in human beings if the fish are ingested. To date experimental evidence has not indicated dangerous levels of accumulation in these biological forms.

Many instances have been reported in the literature of the accumulation of radioisotopes by specific materials, for example, concentration on activated sludges has been reported by Ruchhoft(5), Straub and co-workers(6)(7), Newell and co-workers(8)(9); on trickling filter sludges by Carter(10), Straub and co-workers(6)(11), Newell and co-workers(10), and Butcher(10); on slimes in plumbing fixtures by Reid and Talboys(13); in fish by Knobf(14), Krumholz(15), Higgins(16), and others(17)(18); and on algae by Higgins(16), and Emmons(19). These references will not be reviewed here, but some specific information not readily available in the literature will be included herein.

The data reported by Emmons(19) are of some interest in that an attempt was made to determine what substances were concentrated and to what extent. Tables IV - 2 and IV - 3 contain reported results. It is interesting to note that an apparent concentration of rare earths is indicated for algae.

In studies of radioactivity in fish from White Oak Lake and the Clinch River, Knobf(14) found a relation between activity accumulated and distance from point of discharge of radioactive waste, that is, fish caught in the upper reaches of the White Oak Lake drainage system showed considerably greater activities (423, 2290, and 2901 counts/min/gram of flesh, bone, and scale, respectively) than did fish caught in

TABLE IV - 2

Concentration of Radioactive Materials by Algae$^{(19)}$

Fission Product	Per Cent of Gross Beta Activity Algae	 Water
Sr^{90}-Y^{90}	4.4	92.0
$Sr^{89,90}$	2.4	58.0
Ru^{106}	3.5	<1.0
Ce^{144}	61.0	1.3
Cs^{137}	11.4	<1.0
Per Cent Identified	80.3	~94.0

TABLE IV - 3

Concentration of Radioactive Materials by Algae(19)

Fission Product	Algae c/m/dry gram	Water c/m/ml	Concentration Factor
Sr^{90}-Y^{90}	2.67×10^5	126	2.11×10^3
$Sr^{89,90}$	1.45×10^5	79	1.84×10^3
Ru^{106}	2.09×10^5	2	1.04×10^5
Ce^{144}	3.67×10^6	1.8	2.04×10^6
Cs^{137}	6.85×10^5	2	3.42×10^5
Gross β c/m	6.0×10^6	140	4.33×10^4

the Clinch River several miles below White Oak Creek. At Clinch River mile 13.3 the activities were 2, 13, and 39 counts/min/gram of flesh, bone, and scale, respectively. She also noted that different species of fish accumulated different amounts of activity and that there was considerable variation among individuals of a given species. Reported radiochemical analyses indicated that the activity found in fish bone and scales was due to strontium and yttrium, and in the flesh to cesium (~97 per cent) with the remainder being strontium and yttrium.

A more extensive study of the radioactive materials accumulated by the fish of White Oak Lake is being conducted by Krumholz(15), who has found that there is considerable variation in accumulation in accordance with the seasons. He attributes this variation to increased metabolism with increased temperatures, availability of desirable food, and turbidity of the water. Cesium was found to be the predominant source of activity in the soft tissues and strontium in the harder tissues.

Some unconfirmed preliminary findings in controlled experiments with live gambusia by Cook(20) seem to indicate that in an environment containing Sr^{89}, fish which had died and undergone some disintegration, seemed to have a greater affinity for concentrating radioactive Sr^{89} than did the live fish. These findings are summarized in Table IV - 4.

Summarizing, it may be stated that data available at present on the concentration of radioactive materials by natural agents are of a preliminary nature. There is much to be learned about the many complex mechanisms responsible for this concentration. Interested groups in TVA are carrying out extensive ecological studies in White Oak Creek system to help define some of the factors influencing the reconcentration of radioactive materials by the existing flora and fauna.

TABLE IV - 4

Takeup of Activity (Sr^{89}) from Water by Live and Dead Fish(20)

Fish No.	Temperature of Environment °F	Activity of Environment c/m/ml	Activity - c/m/dry gram*		Concentration Factor	
			Live Fish	Dead Fish	Live Fish	Dead Fish
1	40	4,000	2,800	70,600	---	17.6
2	70	4,000	6,400	130,500	1.6	32.6
3	70	4,000	6,400	195,100	1.6	48.8
4	70	4,000	3,300	50,000	---	15.2
5	40	17,000	16,000	348,300	---	21.8
6	40	17,000	16,000	299,800	---	18.7
7	70	17,000	12,000	314,800	---	26.2
8	70	17,000	12,000	32,000	---	2.7
9	84	17,000	22,000	384,500	1.3	17.5
10	84	17,000	53,200	254,900	3.1	15.0

* Activity in sacrificed weighed whole fish (Gambusia), which were digested with HNO_3 and H_2O_2 before counting.

REFERENCES

1. Cowan, Frederick P. Quantitative Summary of Natural Radiation and Naturally Occurring Isotopes. AECU-1138. U.S. Atomic Energy Commission (January 4, 1951).

2. Morgan, Karl Z. New Public Health Problems in the Atomic Era. Public Health in the World Today, pp. 292-316 (edited by J.S. Simmons) Harvard University Press, Cambridge, Massachusetts (1949).

3. Brockett, T.W., Jr. Unpublished data. Oak Ridge National Laboratory, Oak Ridge, Tennessee (1953).

4. Warren, Shields. Health Problems in Atomic Energy. Digest of Proceedings, Seminar on the Disposal of Radioactive Wastes. Sponsored by the U.S. Atomic Energy Commission, Washington, D.C. (January 24-25, 1949).

5. Ruchhoft, C.C. The Possibilities of Disposal of Radioactive Materials by Biological Treatment Methods. Sewage Works J., 21:5, 877 (September, 1949).

6. Straub, Conrad P. Observations on the Removal of Radioactive Materials from Waste Solutions. Sewage and Industrial Wastes, 23:2, 188 (February, 1951).

7. Straub, Conrad P., Pecsok, Donald A., and Carter, Melvin W. Unpublished data. Oak Ridge National Laboratory, Oak Ridge, Tennessee (1952).

8. Newell, John F. Exploratory Studies on the Removal of Plutonium from Liquid Wastes by Activated Sludge. AECD-2712 (October 4, 1949).

9. Newell, John F., Christenson, C.W., Krieger, H.L., Moeller, D.W., Mathews, E.R., and Ruchhoft, C.C. Laboratory Studies on the Removal of Plutonium from Laundry Wastes. Ind. Engr. Chem. 43:7, 1516 (July, 1951).

10. Carter, Melvin W. Results of an Investigation on the Removal of a Radioactive Isotope (I^{131}) from Sewage by the Use of Laboratory Trickling Filters. Masters Thesis. Georgia Institute of Technology, Atlanta, Georgia (June, 1951). See also Sewage and Industrial Wastes (May, 1953).

11. Straub, Conrad P. Treatment and Disposal of Liquid Radioactive Wastes. Industrial Wastes Treatment. Reinhold Corp., New York, New York (1953).

    (a) Straub, Conrad P., Carter, Melvin W., and Pecsok, Donald A. Unpublished information. Oak Ridge National Laboratory, Oak Ridge, Tennessee (1952).

12. Butcher, Harold E. Removal of Radioactive Wastes by Laboratory Trickling Filters. Masters Thesis. University of Tennessee, Knoxville, Tennessee (1952).

13. Reid, George W. Studies on the Absorption and Assimilation of Radioactive Wastes by Bacterial Slimes on Surfaces. Johns Hopkins University, JHUX-1 (February 15, 1949).

    (a) Studies on the Absorption and Assimilation of Radioactive Wastes by Surface Bacterial Slimes. JHUX-2 (May 15, 1949).

    (b) Informal Progress Report for May 15, 1949 - October 1, 1949. JHUX-3 (October 1, 1949).

    (c) Talboys, Albert P. Retention of I^{131} by Bacterial Slimes in Drains. JHUX-5 (November 15, 1950).

14. Knobf, Venus I. Studies of Radioactivity in Fish from White Oak Lake and Clinch River. Oak Ridge National Laboratory, Oak Ridge, Tennessee. ORNL-1031 (July 9, 1951).

15. Krumholz, Louis A. Unpublished information based on data gathered by TVA under AEC sponsorship. Oak Ridge National Laboratory, Oak Ridge, Tennessee (1952).

16. Higgins, Elmer A. Atomic Radiation Hazards for Fish. J. Wildlife Management, 15:1, 1-12 (January, 1951).

17. Foster, R.F. Effects of Pile Effluent Waters on Columbia River Organisms. 4th Annual Summer Symposium, The Role of Atomic Energy in Agricultural Research. ORNL-ORINS, Oak Ridge, Tennessee (August 25 - 30, 1952).

18. Langford, R.R. A Study of Radioactivity in Ottawa River Organisms and Bottom Deposits. 4th Annual Summer Symposium. The Role of Atomic Energy in Agricultural Research. ORNL-ORINS, Oak Ridge, Tennessee (August 25 - 30, 1952).

19. Emmons, A.H. Unpublished data. Oak Ridge National Laboratory, Oak Ridge, Tennessee (1950).

20. Cook, Mary Jane. Unpublished data. Oak Ridge National Laboratory, Oak Ridge, Tennessee (1953).

SECTION V

WATER DECONTAMINATION

One of the activities receiving major emphasis in the broad program of the Radioactive Liquid Waste Research and Development Section, to which the U. S. Public Health Service and the Corps of Engineers, U. S. Army, have contributed much effort, is a study of water decontamination procedures*. In the studies to be described, an evaluation of existing or conventional water treatment techniques or processes for the removal of radioactive materials from waste solutions has been made. These processes include coagulation, settling, and filtration. In addition, studies have been made of the use of clay (as supplemental turbidity and as a primary method for removing radioactive substances from the waste), phosphate coagulation, ion exchange, metallic dusts, softening, and other procedures.

For the most part, removal efficiencies have been ascertained on a laboratory scale. Pilot plant units have been used to extend the study of coagulation, filtration, and softening processes. Insofar as is known, there are no full-scale water treatment units in use for the removal of specific radioisotopes or mixtures of radioisotopes from water. Studies involving the use of emergency water decontamination procedures are discussed in a separate section following the laboratory and pilot plant investigations.

---

* As stated earlier the findings relative to military types of water treatment are not included because of the classified nature of these studies. See previous citation.

Selection of Radioelements to be Investigated

Water decontamination procedures such as are set forth herein may be applied to the treatment of waters moderately contaminated with radioactive materials. Whether the activity results from wartime or peacetime applications of radioactive materials, the water probably will contain a variety of radioisotopes and it is desirable to know the efficiency of the various treatment processes in removing each of the radioisotopes encountered. In certain instances, a study of a waste containing a mixture of radioelements is feasible as in the use of mixed fission products waste for study of emergency treatment techniques. In other instances, it is more feasible to study radioisotopes individually.

In selecting radioisotopes for study many factors must be considered. Of primary concern is the probability that the radioelement will become a health hazard. This is dependent on the quantity of the material likely to be discharged into the environment and the maximum permissible concentration (MPC) of the material. This criterion limits study to those isotopes which are fission products or which are produced by neutron irradiation of stable isotopes in quantities likely to be hazardous to health. It also limits study to those isotopes of longer half-life and lower MPC value.

The individual isotopes used in studies in this laboratory have been those that are readily available from the Operations Division, ORNL. For convenience in monitoring the activity, it is desirable that the isotopes be beta emitters and have a half-life longer than one day. A further limiting factor is cost. The radioelements which can be investigated with these criteria in mind are tabulated in Table V - 1 according to the Periodic System.

Of the 46 elements listed in Table V - 1, 26 are found in fission products, namely: zinc through europium. These 26 elements include those radioelements of maximum health hazard following a bomb blast with the possible exception of tellurium and zenon. They also include those fission products of half-life greater than one year with the exception of krypton. The 20 other elements listed in Table V - 1 have radioisotopes which can be induced by neutron bombardment and include those likely to be released into watercourses in peacetime application. Radioisotopes of the underlined elements in Table V - 1 have been studied by this laboratory in some degree, and at least one element in each Periodic Group is represented.

TABLE V - 1

Radioelements Suitable for Study **

Period	Group								
	0	1	2	3	4	5	6	7	8
1		H							
2					C				
3						$\underline{\text{P}}$	S	Cl	
4			Ca $\underline{\text{Zn}}$	$\underline{\text{Sc}}$		As	$\underline{\text{Cr}}$ Se	Br	Fe $\underline{\text{Co}}$ Ni
5		Rb Ag	$\underline{\text{Sr}}$ $\underline{\text{Cd}}$	$\underline{\text{Y}}$ In	$\underline{\text{Zr}}$	$\underline{\text{Nb}}$ $\underline{\text{Sb}}$	Mo	$\underline{\text{I}}$	$\underline{\text{Ru}}$ Rh
6		$\underline{\text{Cs}}$ Au	$\underline{\text{Ba}}$ Hg	La* Tl		Ta Bi	$\underline{\text{W}}$	Re	Os Ir
7									
6	Rare earths *La series - $\underline{\text{La}}$ $\underline{\text{Ce}}$ $\underline{\text{Pr}}$ Nd Pm Sm Eu								

** Underlined elements have been investigated by the Radioactive Liquid Waste Research and Development Section, Health Physics Division, ORNL.

Laboratory Studies

Procedure

Standardized jar test procedures were employed to evaluate the removal efficiency of coagulation and softening processes, addition of turbidity (clay), and metallic dusts. In the filtration and ion exchange studies, small columns of bed material were used.

For the jar tests, specific concentrations of coagulating chemicals, radioisotopes, and/or turbidity were added to a given quantity of Oak Ridge tap water*. In general, 500 ml of solution were placed in one-liter beakers and subjected to a three-minute rapid mix (three feet per minute peripheral velocity) and a 27-minute slow mix (0.7 feet per minute peripheral velocity). After mixing, the solutions were allowed to settle for 30 minutes to two hours. Depending on the concentration of radioactive materials present, various volumes of the initial liquid, supernatant liquid after treatment, and settled sludge were extracted, placed in aluminum counting dishes, dried under infra-red heat lamps, and counted in a Geiger-Mueller end-window counter (geometry approximately 10 per cent).

For the column studies, 50 ml burettes or columns of similar size with a bed depth of about 18 inches were employed, and the flow rates utilized were those comparable to rapid sand filter rates (about 125 mgad) used in conventional water treatment practice. Samples of the effluent were collected at regular time intervals and the counting procedure was the same as that above.

---

* Average analyses of Oak Ridge tap water will be found in Appendix "B".

All individual radioisotopes used in these studies were procured from the Operations Division, ORNL, and were in the following chemical forms: P^{32} initially present as phosphate; $Ce^{141, 144}$ - Pr^{144}, Sr^{89}, Sr^{90} - Y^{90}, Y^{91}, Sb^{124}, Ru^{106}, Cs^{137}, Ba^{140} - La^{140}, Sc^{46}, Zn^{65}, Cr^{51}, and Co^{60} as chlorides; I^{131} as the iodide; Zr^{95} - Nb^{95} as the oxalate complex in acid solution; W^{185} as the tungstate; and Cd^{115} as the nitrate. Several mixed fission product wastes were also investigated.

Addition of Clay Particles (Turbidity)

Surface waters contain various amounts of turbidity, usually in the form of clay or fine silt particles. Some radioisotopes, either through adsorption, ion exchange, or a combination of both, will be removed by this natural turbidity. In a recent publication(1) Freundlich-type isotherms, which showed the relation between the amount of activity adsorbed per unit of adsorbent and the concentration of activity remaining, were plotted for several radioisotopes. On the basis of the results shown in these plots, the assumption was made that the mechanism involved in the removal of these isotopes was primarily adsorption.

Studies have been performed in which the addition of clay alone followed by sedimentation has been the primary means of treatment. Jar test experiments were made using various doses of local clay with a number of isotopes. The initial activities of the solutions treated were in the order of 10^{-2} uc/cc. Representative data for three clay concentrations are given in Table V - 2. These data show wide variability in removal of the elements studied. In some cases the data have been shown to follow a Freundlich-type isotherm, or, in other words, for a given concentration of contaminant the removal per unit of adsorbent is much greater at lower concentrations of clay than at higher concentrations of clay.

Lauderdale investigated the removal of radioisotopes from diluted pond water containing mixed fission products by the use of montmorillonite and kaolinite clay(2). These studies illustrate the variation in removal due to the particular clay used. His data have been plotted utilizing the Freundlich-type isotherm. To adapt the Freundlich-type equation to the adsorption of radioisotopes, the equation is written in the form:

$$x/M = KC^n \text{ or } \log x/M = \log K + n \log C$$

TABLE V - 2

Effect of Turbidity* on Removal of Radioisotopes

Radioisotope	Per Cent Removal 100 ppm	750 ppm	5000 ppm
Ba^{140}-La^{140}	41	58	85
Cd^{115}	3	30	64
Ce^{144}	70-80	86	--
Cs^{137}	38	87	98.0
Sc^{46}	53	91.7	96.9
Sr^{89}	2-12	14-22	49-52
W^{185}	0	4	49
Y^{91}	22-45	56-70	93.6
Zr^{95}-Nb^{95}	93.5	99.0	98.0

* Local clay used to produce turbidity

Note: In the reporting of results, the average value is given. Where two or more comparable studies have been performed. the range in averages for studies is reported. Removals less than 90 per cent are given to the nearest integer per cent. Removals exceeding 90 per cent are given to the nearest tenth of a per cent.

where,

M = concentration of clay added (ppm)

x = concentration of activity removed or C_o - C where C_o is the initial activity and C the final activity (cts/min/ml)

K and n are experimental constants

It was found that the kaolinite clay exhibited higher removal efficiencies at low initial counts whereas at high initial counts (above 10,000 c/m/ml) the montmorillonite clay appears more efficient. For example, with an initial activity of about 1,400 counts per minute per ml (c/m/ml), the amount of montmorillonite clay required to effect a 60 per cent reduction in activity is approximately 20 times the amount of kaolinite clay required to effect the same reduction. To obtain the same percentage activity removal at an initial count of 14,000 c/m/ml, the amount of montmorillonite clay required is approximately one-half that of the kaolinite clay.

Various amounts of bentonitic montmorillonite, kaolinite, and montmorillonite clays were added to a fission product mixture which had an initial concentration of radioactive material in the range of 1,385 to 1,485 cts/min/ml. At the lower clay concentrations kaolinite clay removed greater fractions of the radioactive constituents than did similar additions of the other two types of clay.

With the bentonitic montmorillonite and montmorillonite clays stable salts are apparently preferentially removed and consequently, greater concentrations of these clays are required to effect the same removal of radioactive contaminants as the kaolinite clay. It may be inferred from these studies that increasing concentrations of stable salts in the water are deleterious to removal of radioactive contaminants.

Studies have been made of the removal of mixed fission products from water using several sources of waste and several types of clays. Removals varied widely and depended on the chemical and radiochemical composition of the waste in addition to the type and concentration of turbidity added.

Studies with Ce^{144} and Y^{91} have shown that removals increased with increased pH values(3). For example, in the case of Ce^{144}, the removal in the pH range of 4 to 8 was about 65 per cent, and was increased to about 98 per cent at pH 11. With Y^{91} somewhat similar findings were reported with removals of about 70 per cent in the pH range 5 to 9 and above 90 per cent at pH 11. More recent studies(4) with Co^{60}, Ir^{192}, and Tl^{204} show very definite increases in removal at increasing pH values. In general, it may be stated that higher removals are associated with higher pH values. In past studies(3), the rate of mixing was found to be significant. With five-minute rapid mix followed by 25-minute slow mix (same peripheral velocities as indicated earlier) 25 per cent of the Ce^{144} and 39 per cent Y^{91}

were removed from solution. In another series in which 30-minute rapid mix was utilized the removals for these two radioisotopes were 93 and 94 per cent respectively.

Summarizing, the extent of removal by clay or turbidity of radioisotopes from waste solutions varies over a wide range and is primarily dependent upon: the radioisotope involved and its concentration, the concentration and type of clay present, the composition of the substrate solution including the concentrations of carrier and other salts, the pH of the solution, and the rate of mix or time of contact. Extreme caution should be exercised, therefore, in interpreting general statements that have been made on overall removals of radioactive contaminants by suspended or colloidal matter.

From a practical standpoint in the treatment of large volumes of water, the addition of clay in excess of 100-500 ppm results in a tremendous materials handling problem of both raw material and resultant sludge. For clay additions of 100 to 500 ppm, the removals of mixed fission products from low-activity waters would be roughly 25 to 50 per cent. For example, for a waste containing 10^{-2} uc/cc, such treatment might produce an effluent with an activity in the order of 5×10^{-3} uc/cc. Consequently, the addition of clay probably would be used in conjunction with some other decontamination process.

Of the individual radioelements studied, the use of clay would be particularly advantageous for treatment of a waste containing significant amounts of radioactive cesium. Other water treatment procedures with the exception of ion exchange have shown negligible removals of this element whereas removals in the order of 90 to 98 per cent can be effected with addition of 1,000 to 5,000 ppm clay.

Coagulation and Settling

The results obtained in routine coagulation studies for the removal of Ce^{144}, Y^{91}, Sr^{89}, I^{131}, P^{32}, and iodine dissolver solution (a fission products mixture) by different coagulants under various conditions of dosage have been reported in detail(1). Since these studies were reported, additional data have been obtained with Cs^{137}, Cd^{115}, Zr^{95} - Nb^{95}, W^{185}, Sr^{89}, Sc^{46}, Cr^{51}, Y^{91}, an equilibrium mixture of Ba^{140} - La^{140}, and a fission products mixture which was assumed to simulate a 30-day old bomb blast mixture. The studies with tungsten, strontium, scandium, chromium, yttrium, and barium-lanthanum were extensive investigations utilizing three coagulants ($FeSO_4$, $FeCl_3$, and alum), three coagulant dosages (1, 2, and 6 gpg*) and three activity levels (about 20,000 counts per minute per ml (c/m/ml), 2,000 c/m/ml, and 200 c/m/ml). All jar test results, where coagulation with alum or iron salts was utilized, are summarized in Table V-3. Wide variations in removal efficiencies are indicated and certain general statements can be made regarding these variations.

---

* Unless otherwise noted, a specified coagulant dosage level defines the amount of coagulant, alkaline reagent, and sodium silicate added. In each case, the coagulant and alkaline reagent dosages are the value indicated whereas the amount of silicate added is 40 per cent of the coagulant dose. For example, a 2 gpg coagulant dosage represents 2 gpg alum (as $Al_2(SO_4)_3 \cdot 18\ H_2O$) or ferrous sulfate as ($FeSO_4 \cdot 7\ H_2O$) or ferric chloride (as $FeCl_3 \cdot 6\ H_2O$); 2 gpg lime (as $Ca(OH)_2$) or soda ash (as Na_2CO_3) or sodium hydroxide (as $NaOH$); and 0.8 gpg sodium silicate (as SiO_2).

(1) Greater removals are effected when turbidity is initially present; or, to some extent the removals gained by coagulation and by turbidity may be additive.

(2) Removals increase with increasing coagulant dose; however, the unit gain of removal by a given coagulant rapidly diminishes with increasing dosages.

(3) The three coagulants studied (alum, $FeSO_4$, and $FeCl_3$) effect removal efficiencies of the same order in the normal ranges of pH (pH 6 to 8). The iron salts gave somewhat higher removals than alum for water contaminated with tungsten, strontium, and yttrium; however, these higher removals were associated with higher pH levels where the broader pH range of the iron salts undoubtedly aided removal. Coagulation with $FeCl_3$ was somewhat inferior to either alum or $FeSO_4$ in the removal of scandium. Comparison of the removal efficiencies of the three coagulants (in the case of chromium, barium-lanthanum, cerium, and iodine) indicated practically no difference in removal efficiency. The use of iron salts rather than alum results in reduced sludge volumes. For example, if 6 gpg coagulant are added

TABLE V - 3

Coagulation and Settling Results - Jar Test Studies

Isotope	Turbidity Added (ppm)[a]	Coagulant Added (gpg)[b]	Final pH	Per Cent Removal
Ce^{144}	0	1 - 1.5*	7.2- 7.8	81-94
	100	0.5 - 2.5*	7.0- 7.8	85-96
Y^{91}	0	1.5*	6.8- 7.1	83-93
	100	1 - 6	7.0-10.2	34-99
Sr[illegible]	0	1.5*	6.7- 7.8	0- 6
	100	0.5 - 6	6.7-10.7	0-51
Zr^{95}-Nb^{95}	0	1 - 5	-	70-98
	100	1	-	95-99
Cs^{137}	0	1	-	0- 6
	100	1	-	35-65
Cd^{115}	0	1	-	40-60
	100	1 - 5	-	60-95
W^{185}	100	1 - 6	7.5- 8.4	5-91
Sc^{46}	100	1 - 6	6.5- 8.2	66-98
Cr^{51}	100	1 - 6	7.6- 8.8	73-98
Ba^{140}-La^{140}	100	1 - 6	7.5- 8.2	28-84
I^{131}	100	0.5 - 2*	6.9- 9.0	0-10
P^{32}	100	0.5 - 1.5*	6.8- 8.8	97-99
FPM-1	100	1 - 3*	7.2- 8.8	61-84
FPM-2	0	1 - 5	4.3-10.2	9-71
	100	1 - 5	4.3-10.2	12-73
FPM-3	0	10	9.9-10.0	46

[a] Local clay was added for turbidity

[b] Coagulant includes alum, ferrous sulfate, or ferric chloride, lime, soda ash, or sodium hydroxide, and sodium silicate.

* No sodium silicate added.

FPM-1 - Iodine dissolver solution.

FPM-2 - Synthetic mixture containing fission products in the same concentrations assumed to be present 30 days after an under water bomb blast.

FPM-3 - Three year old fission product mixture.

to a waste containing 100 ppm turbidity, the sludge volumes after two hours settling amount to about 1.2 per cent for the iron salts and about 2.5 per cent for alum. The volume of alum sludge is dependent upon the final pH of the waste under investigation.

(4) The use of either soda ash, lime, or sodium hydroxide as the alkaline reagent gave negligible differences in removal efficiency aside from the differences concommitant to changes in pH. In this regard, certain data are conflicting particularly with yttrium, but where the data are better substantiated, it appears that final pH rather than type of alkaline reagent is the more pertinent variable. This has been demonstrated with Sr^{89} and Y^{91} where, with pH values ranging from about 7 to 10, removals for strontium increase uniformly from about 10 to 50 per cent with increasing pH and removals for yttrium increase from about 50 per cent to greater than 95 per cent. The pH range in studies with other individual radioisotopes has not been sufficiently broad to show whether or not this same phenomenon is operative. It was conclusively shown, however, for a fission products mixture (FPM - 2, Table V - 3), that substantially greater removals are obtained in an alkaline medium, but no appreciable differences in removal were noted at pH ranges of about 7.5 and 10. Where high pH values increase removals, the use of lime or sodium hydroxide would be preferable to soda ash.

(5) In general, 100-fold variations in activity level, i. e., 200 c/m/ml to 20,000 c/m/ml, did not have a consistent effect on percentage removal values of isotopes studied. In one instance, that of tungsten, removals increased somewhat at lower activity levels whereas with a fission product mixture (FPM - 2) the reverse effect was shown. Carrier additions up to 1 ppm of strontium with Sr^{89}, and carrier additions of zirconium and niobium up to 0.01 ppm with Zr^{95} - Nb^{95} had a negligible effect on removal efficiencies.

Certain specific comments regarding the removal of particular radioisotopes may be made as follows:

(1) Under the conditions employed chemical coagulation effected only slight removals of Sr^{89}, Cs^{137}, and I^{131}. The removal of I^{131} can be enhanced by addition of $AgNO_3$ or activated carbon; for removals in the order of 75 per cent 4 ppm $AgNO_3$ or 5 - 15 ppm carbon may be required.

(2) With the coagulation procedures used the maximum removals obtained from mixed fission product wastes (noted in Table V - 3) are in the order of 40 to 70 per cent and the addition of 100 ppm of turbidity increased removals to 50 to 80 per cent.

(3) Studies with Ba^{140} - La^{140} using step coagulation and sludge return showed negligible effect on removal efficiency. In a step coagulation experiment, 1 gpg each of alum, $FeSO_4$ and $FeCl_3$ were added in six successive steps and the overall removals were

approximately the same as a single 6 gpg dosage of these coagulants. In sludge return studies, solutions containing Ba^{140} - La^{140} were coagulated with the same three coagulants whereupon the sludge was returned to a fresh solution along with the same initial dosage of coagulants. Four such return passes were made with no beneficial results due to sludge return.

Eliassen and co-workers(5) have reported laboratory studies on the removal of P^{31} + P^{32}, I^{131}, and Sr^{89} with results in agreement with those indicated above. In a discussion of this paper, Swope(6) included data on the removal of mixed beta emitters from waste solutions using lime, $FeCl_3$, silica product, and NaOH, either singly or in combination. Removals were reported to be 80 to 92 per cent. Her data are somewhat unusual in that the lower removals were obtained at higher pH values (10.7 to 11.1), and the higher removals (89 to 92 per cent) were associated with lower pH values (8.1 to 9.6).

Studies in which 50 and 100 ppm, respectively, of ferric chloride and limestone or 40 and 200 ppm, respectively, of alum and limestone were used for the removal of I^{131}, $Ce^{141, 144}$ - Pr^{144}, Ba^{140} - La^{140}, Zr^{95} - Nb^{95}, Co^{60}, and two fission products mixtures have been described by Lacy(7). His findings are summarized in Tables V - 4 and V - 5.

TABLE V - 4

Ferric Chloride-Limestone Coagulation Results(7)
(30-minute stirring followed by 60-minute settling)

Contaminant	Initial pH	Initial Activity c/m/ml	$FeCl_3$ ppm	$CaCO_3$ ppm	Removal Per Cent
I^{131}	7.7	2,480	50	200	45.0
			100	200	63.0
$Ce^{141,144}$-Pr^{144}	7.6	4,100	50	200	99.2
			100	200	99.4
Ba^{140}-La^{140}	7.7	4,620	50	50	67.4
			100	100	70.7
Co^{60}	8.2	4,590	50	150	22.9
Zr^{95}-Nb^{95}	8.7	2,980	50	200	98.1
			100	400	98.8
FPM-4a	8.2	590	50	100	32.0
			100	200	40.1
FPM-4	10.0	10,000	170	240	88.7
FPM-5	8.5	2,220	50	200	51.1
			100	400	59.4

FPM-4 - Iodine dissolver solution aged 30 days.
FPM-4a- FPM-4 pretreated with clay.
FPM-5 - Mixed fission product waste containing mainly Cs^{137}-Ba^{137} and Ru^{106}-Rh^{106}.

TABLE V - 5

Alum and Limestone Coagulation Results(7)
(30-minute stirring followed by 60-minute settling)

Contaminant	Initial pH	Initial Activity c/m/ml	Alum ppm	$CaCO_3$ ppm	Removal Per Cent
I^{131}	6.3	1,300	100	100	26.3
			200	200	35.7
$Ce^{141,144}$-Pr^{144}	7.1	5,580	50	200	96.1
			100	200	96.5
Ba^{140}-La^{140}	7.8	1,750	50	50	58.4
			100	100	58.0
Co^{60}	7.6	4,940	50	100	8.2
Zr^{95}-Nb^{95}	8.4	7,800	40	40	76.4
			80	80	78.6

From an examination of these results, it will be seen that when ferric chloride and limestone were used removals of $Ce^{141, 144}$ - Pr^{144} and Zr^{95} - Nb^{95} greater than 90 per cent were obtained, a removal of about 60 to 70 per cent with Ba^{140}- La^{140}, a removal of 45 per cent with I^{131}, and that lower removals were noted with Co^{60}. On the basis of the results, it appears that the iron-limestone combination was more effective than the alum-limestone combination. However, the results were inconclusive since the amount of coagulating chemicals was varied and the initial pH was not constant when using the two coagulants.

The results of coagulation studies reported for the removal of plutonium from waste solutions at Los Alamos(8) may be summarized as follows:

(1) Coagulation with 20 ppm $FeCl_3$ and sufficient lime to adjust the pH to 9.5 resulted in removal of 99.28 per cent of the initial activity of plutonium from laundry wastes. The initial activity averaged 1,815 c/m/liter and the final activity averaged 13 c/m/l which is below the permissible level of 70 c/m/liter set for plutonium. About 20 gallons of wet sludge were produced per 1,000 gallons of waste solution resulting in about three pounds of dry solids.

(2) Complexing agents such as citrates and phosphates interferred with the coagulation procedures. The interference was over-

come by addition of calcium ions and by increasing the pH to about 12. Activated sodium silicate is valuable in assisting floc formation.

Coagulation with alum or iron salts is the principal means by which the average water treatment plant, without modification, could attempt decontamination of a water supply. If a moderate amount of turbidity ($\sim$ 100 ppm) is assumed normally present and if a coagulant dosage of about 6 gpg is attainable with standard chemical feeding equipment, the average treatment plant could effect a removal of mixed fission products from water in the range of 25 to 75 per cent. The addition of clay (in practicable amounts up to 500 ppm), the addition of alkali to raise the pH, or the addition of further amounts of coagulant would enhance removal of radioisotopes but decontamination of water containing mixed fission products would probably not exceed 90 per cent with these measures. If the 10-day emergency MPC value of 9×10^{-2} uc/cc is considered, the raw water could not contain more than 1 uc/cc if a satisfactory water is to be produced with modified standard water treatment processes.

Sand Filtration

Studies with individual isotopes have been performed at ORNL pertaining to the removal of radioactive mamerials by laboratory-scale rapid sand filters. In these studies, 500 or 750 ml of tap water solution containing a particular isotope was passed through a sand bed contained in a 50 ml burette. The size of the washed sand and the rate of flow were comparable to rapid sand filtration practice. The results are reported in Table V - 6.

Referring to Table V-6, removals in the order of 90 per cent were effected for yttrium, scandium and zirconium-niobium. These three isotopes are believed to be in colloidal form and, consequently, are probably removed by true filtration. With the other isotopes studied, it is likely that a limited degree of adsorption takes place but in each instance, removals are negligible in view of the rapid increase of radioactivity in the effluent.

In conjunction with conventional water treatment, sand filtration following coagulation and sedimentation would remove some radioactive contaminants. However, the mechanism of removal would be the filtering out of fine floc particles (not removed by settling) which have occluded the contaminants.

Lime-Soda Softening

Studies of lime-soda softening at ORNL have included work with Ba^{140} - La^{140}, Sr^{89}, Y^{91}, W^{185}, Sc^{46}, Zr^{95} - Nb^{95}, Cd^{115}, and Cs^{137}. This summary also includes the results of studies with Sr^{89} - Y^{90} by W. A. Hoyt(9) and with Sr^{89} by E. F. Gloyna(10). Removal efficien-

TABLE V - 6

Laboratory Sand Filtration Results

Isotope	Initial Activity c/m/ml	Volume Passed ml	pH of Effluent	Per Cent Removal Range	Average
Ba^{140}-La^{140}	1,300	750	7.6	39-99	74
Cd^{115}	1,200	500	8.1	60-99	95
Cs^{137}	800	500	8.3	10-70	50
Sc^{46}	1,500	750	8.3	94-99	96
Sr^{89}	2,700	750	8.3	1-13	4
W^{185}	2,200	750	7.1	3-18	8
Y^{91}	5,700	750	7.0	84-89	87
Zr^{95}-Nb^{95}	3,400	500	7.8	91-96	93

cies for these isotopes were determined using variable excess dosages of lime and of soda ash.

In general, the procedure was similar to that used for coagulation experiments, i. e., the radioisotopes were added to Oak Ridge tap water followed by variable amounts of lime and soda, and final samples obtained after 1 minute rapid mix, 30 minute slow mix and 2 hours settling. Hoyt used White Oak Creek water instead of tap water and Gloyna used a modified distilled water.

Results using tap water are summarized in Table V-7. The stoichiometric dosages for treatment of this water are approximately 6 gpg lime (as $Ca(OH)_2$) and 1 gpg soda ash (as Na_2CO_3). The results for the isotopes studied indicate that excess treatment would be required for removals of 90 per cent or greater.

To analyze these results more completely, the isotopes studied will be grouped according to the Periodic System and the groups will be discussed individually.

Barium, Strontium, and Cadmium (Group II Elements) - As indicated in Table V-7, cadmium is removed most readily, followed by barium-lanthanum. Strontium required somewhat higher dosages of lime and soda ash to give similar percentage removals. Since removal of strontium is more pertinent from a health hazard standpoint, certain additional comments regarding strontium are made as follows:

TABLE V - 7

Approximate Minimum Combined Dosage* of Lime and Soda Ash to Give Stated Removal

Isotope	Chemical Dosage in gpg for Stated Per Cent Removal									
	50		75		90		95		99	
	Lime	Soda	Lime	Soda	Lime	Soda	Lime	Soda	Lime	Soda
Ba^{140}-La^{140}	2	2	4	2	6	4				
Sr^{89}	4	3	5	5	7	9	20	20		
Cd^{115}	2	2	3	3	4	4	4	4	6	4
Sc^{46}	3	3	3	3	5	5				
Y^{91}	2	2	4	4	6	6	12	6		
Zr^{95}-Nb^{95}	2	0	5	0	12	0	17	0	22	0
Cs^{137}	48	48								
W^{185}	48	48								

* Minimum combined dosage is defined such that, of those variable dosages studied, the gpg lime plus the gpg soda ash is a minimum.

(1) To obtain removals in the order of 95-99 per cent, the results given in Table V-7 confirmed Hoyt's findings that dosage of at least 20 gpg lime and 20 gpg soda ash are required. Similar results were obtained in the study of a mixed radioactive waste containing about 75 per cent Sr^{89} (in terms of activity).

(2) Other modifications of lime-soda treatment studied by Hoyt were in no way as effective as excess treatment. For example, using a stoichiometric softening dose, which gave 75 per cent removal of Sr^{89}, the addition of 300 ppm $Sr(NO_3)_2$ increased removal to about 95 per cent. With the same softening dose, raising the pH level from 10.5 to 12 increased removal to about 83 per cent. The addition of ferric chloride coagulant had negligible effect on removal. Softening in two steps (such as two additions of 100 ppm excess lime-soda ash) as compared with a single addition (200 ppm) showed negligible increase in removal efficiency. In other ORNL studies, recarbonation of the supernatants of excess treated samples showed no appreciable further increase in removals.

(3) The effect of carrier was studied by Gloyna who added Sr^{89} and 100 ppm $Sr(NO_3)_2$ to distilled water modified by adding NaOH to a pH of 12. Subsequent stirring of this solution allowed precipitation of $SrCO_3$ with a resulting 99.8 per cent reduction in activity. Application of such treatment where $SrCO_3$ is precipitated

is impractical from a cost standpoint.

(4) The mechanism of removal of strontium with excess lime treatment is most likely coprecipitation by means of mixed crystal formation. Such contention is borne out by the fact that cadmium with an ionic radius similar to calcium is more readily removed than barium or strontium which have larger ionic radii.

Yttrium and Scandium (Group III Elements)

These elements are removed quite easily with comparatively small dosages (about equal to the stoichiometric dose) effecting removals in excess of 90 per cent.

Zirconium-Niobium (Groups IV and V)

Removal of these elements can be attained by lime additions alone which indicates that a high pH level aids the precipitation of colloidal zirconium and niobium.

Cesium (Group I) and Tungsten (Group VI)

Practically no removal of these elements resulted with dosages of softening reagents as high as 48 gpg.

In summary, lime-soda softening is fairly effective in removing certain radioelements if relatively high chemical dosages are employed. From the standpoint of emergency water treatment, this process is probably of little concern in that waterworks serving less than 15 per cent of this country's population are presently equipped to soften water Also, exceedingly high dosages would be required to effect substantial reduction of mixed fission products. The softening process could be adapted to treatment of a waste containing radio-strontium. Other water treatment processes, with the exception of ion-exchange, are relatively ineffective in removing this particular element whereas softening, under optimum conditions, can effect removals in excess of 99 per cent.

Ion Exchange

The most extensive studies thus far reported in the literature on the use of ion exchange for the removal of radioactive constituents on a laboratory scale are those by Ayres(11). He concludes that ion exchange treatment of radioactive waste is feasible. He further indicates that since the primary purpose of treating radioactive wastes is to effect a volume reduction it would be impractical to regenerate the resins for reuse. Three ion exchange methods which may be used, each having its own particular advantages are: two-bed deionization, monobed deionization, and cation exchange. Although the discussion of these methods is restricted to removal of radioactive materials from waste solutions, the methods may be applied to removal of radioactive constituents from water to be used for drinking and culinary purposes. Ayres' description of these methods is as follows:

Two-bed Deionization - In this method two columns would be required, one for the cation and one for the anion exchange resin. The liquid waste would flow first through the cation exchanger, then through the anion exchanger, and finally be discharged. After depletion, the resin would be removed and incinerated. The decontamination factor would be in the order of 10^5.

Monobed Deionization - Although the cost and the concentration and decontamination factors would be the same as for the conventional two-bed system, there might be a slight advantage in ease of operation and size of installation.

Cation Exchange - Where known isotopes are used which can be removed by cation exchange materials, and where a high decontamination factor may not be necessary, cation exchange columns are quite satisfactory. For example, some laboratories may use only small amounts of radioactive tracers and the activity may be diluted by the other wastes. In this case, a simple cation exchange column may be sufficient since it would remove some of the most troublesome radioactive isotopes such as barium, strontium, zirconium, niobium, and the rare earths. All anions such as iodine pass on through, and the amphoteric elements, such as molybdenum and ruthenium, are removed only in part. The apparatus would consist of a single column filled with cation exchange resin and provision could be made to neutralize the effluent.

Ayres(11) also studied the effect of ordinary contaminants such as oils, solvents, and precipitating and complexing agents on removal of radioisotopes by ion exchange columns and concludes that these contaminants do not have a deleterious effect upon removal.

Some work with ion exchange resins has been carried out by the RLWRDS at ORNL and these results are summarized in Table V - 8. In these studies, approximately 40 ml of ion exchange material was placed in a 50 ml burette and tap water solutions of the radioisotope were passed through the column at the rate of 2 gallons per square foot per minute. From 500 to 750 ml of solution were passed through the columns and in no case was the ion exchange material exhausted with regard to hardness or activity. The purpose of the tests was to determine whether a particular radioisotope would be removed by ion exchange. No attempt was made to estimate the capacity of any of the materials for a given radioisotope.

The results shown in Table V - 8 are average removal values for the total volumes of solution passed through the columns. The cation exchange materials evaluated included IR-120 on the H^+ and

TABLE V - 8

Percentage Removal Results of Individual Isotopes with Ion Exchange Materials

Isotope	Ion Exchange Material Cation Ex	Anion Ex	Mixed Bed	Greensand
W^{185}	12 -16	97.2-99.2	98.9	9
Y^{91}	86 -93.1	94.2-98.5	97.6 -98.7	75
Sc^{46}	95.7-97.2	98.8-99.0	98.5 -98.7	96.4
Sr^{89}	99.1-99.8	5 - 7	99.95-99.97	99.8
Ba^{140}-La^{140}	98.3-99.0	36 -42	99.5 -99.6	96.3
Cs^{137}	99.8	9	99.8	--
Cd^{115}	98.5	0	99.2	--
Zr^{95}-Nb^{95}	58 -75	96.4-99.9	90.9 -99.4	--

Na^+ cycles; the anion exchangers included Dowex - 1 on the OH^- and Cl^- cycles; and the mixed beds included an IR-120 (H^+) - Dowex - 1 (OH^-) bed and an IR-120 (Na^+) - Dowex - 1 (Cl^-) bed. A natural greensand was also evaluated for comparison with these resins. In general, the particular regeneration cycle, i. e., whether H^+ or Na^+ regenerated cation resin or OH^- or Cl^- regenerated anion resin, did not affect the results appreciably. In certain applications, the effluent pH may be of significance and the range in average effluent pH's for the various materials is as follows: IR-120 (H^+), 2.8 - 3.6; IR-120 (Na^+), 3.4 - 4.3; Dowex - 1 (OH^-), 10.7 - 11.2; Dowex - 1 (Cl^-), 7.2 - 8.7; Greensand, 7.1 - 8.0. The removal efficiencies with greensand do not appear to be significantly different from the sodium-cycle cation exchange resin; however, their total capacities undoubtedly differ greatly.

It will be noted that strontium, cesium, cadmium, and to a lesser extent barium-lanthanum are present in solution as cations, whereas tungsten is present as an anion. Yttrium, scandium, and zirconium-niobium are apparently in colloidal form and, as such, are removed to some extent on both cation and anion resins. In this regard Ayres(11) cites early work on the Manhattan Project which indicated that colloids in radiochemical concentrations, especially zirconium and niobium when passed through exchange columns were removed by adsorption on the resin rather than by ion exchange. It was stated that "this was not ion exchange since once adsorbed, they (zirconium and niobium) could not be removed from the column by solutions containing high concentrations of ionized salts. It was necessary to use some complexing agent such as dilute oxalic acid as an elutant."

A more extended study of ion exchange was made by Friend(12) who investigated this procedure for the removal of I^{131}, Sr^{89}, and mixtures of I^{131} and Sr^{89} from Oak Ridge tap water dilutions of these radioisotopes. A strongly basic type anion resin (IRA-400) was used on the Cl^- and OH^- cycles and a strongly acid type cation resin (IR-120) was used on the H^+, Na^+, and the Ca^{++} cycles. The investigation was conducted in three parts: (1) the removal of carrier-free I^{131} by the anion resin and mixed resin bed; (2) the removal of carrier-free Sr^{89} by the cation resin and a mixed resin bed; and (3) the removal of a mixture of carrier-free I^{131} and Sr^{89} by a mixed resin bed. The mixed bed in each instance contained equivalent amounts of both resins. A decontamination factor (influent activity divided by effluent activity) in excess of 10^2 was obtained for carrier-free I^{131} through the use of IRA-400 resin on the OH^- cycle or a combination of IR-120 (Na^+ or H^+ cycles) and IRA-400 (OH^- cycle); a decontamination factor in excess of 10^3 for carrier-free Sr^{89} through the use of IR-120 on the H^+ cycle or a combination of IR-120 (H^+ or Na^+ cycles) and IRA-400 (OH^- cycle); and a decontamination factor in excess of 10^3 for the carrier-free mixture of Sr^{89} and I^{131} through the use of a mixture of IR-120 (H^+ or Na^+ cycles) and IRA-400 (OH^- cycle). The initial activities used in these tests were approximately 2×10^{-2} uc/cc; the activities remaining, in the order of 10^{-4} to 10^{-5} uc/cc.

Studies describing the use of mixed cation and anion resins for the removal of a variety of radioisotopes from water solutions have been reported by Lacy(7). In these studies, varying concentrations of a combined cation-anion resin (Aberlite MB-3) were added to solutions containing specific radioisotopes. These mixtures were stirred by means of a laboratory stirring device and the takeup of the radioactive contaminant on the resin evaluated by measuring the residual radioactivity in the supernatant liquor after 90 minutes settling, and filtration. Removals of 98.1 to 99.99 per cent are reported when 2,700 ppm of resin were used. The results of these studies are summarized in Table V - 9.

Although the ion exchange process offers one of the most promising methods for removing radio-contaminants, the cost of the resins may preclude widespread usage unless suitable regeneration techniques are developed. In some cases, through the proper choice of regenerant, it may be possible to elute the stable ions from the resin while retaining the radioactive material. Normally, regener-

TABLE V - 9

Removal of Radioactive Contaminants from Water
by Combined Anion-Cation Resin(7)
Resin-Jar Test Studies (Stirring time 90 minutes, samples filtered)

Contaminant	Initial pH	Initial Activity c/m/ml	Per Cent Removal Mixed Ion Echange Resin - ppm 450	900	1800	2700
P^{32}	8.2	5,560	47.4	74.5	96.2	99.8
Cd^{115}	8.0	7,880	37.9	45.6	91.1	99.99
Cs^{137}-Ba^{137}	8.2	8,200	15.1	14.6	69.1	99.99
Zr^{95}-Nb^{95}	8.1	6,700	98.3	98.4	99.2	99.4
I^{131}	7.5	3,200	84.5	93.5	95.6	98.1
$Ce^{141,144}$-Pr^{144}	7.9	4,150	98.7	99.2	99.8	99.98
Ba^{140}-La^{140}	7.6	3,490	85.1	94.5	98.8	99.9
FPM-4	8.3	13,600	82.7	90.5	97.3	99.2
FPM-5	2.7	3,400	38.4	--	--	--

FPM-4 - Iodine dissolver solution aged 30 days.
FPM-5 - Mixed fission product waste containing mainly Cs^{137}-Ba^{137} and Ru^{106}-Rh^{106}.

ation elutes the resin completely and techniques are required such that the regenerant volume is kept to a minimum. Studies are under way at ORNL and at the University of California utilizing a continuous regenerating method which may eliminate some of the objectionable features which result from single-pass ion exchange procedures.

Summarizing, when radioisotopes are in an ionized form, it is possible to obtain a decontamination factor of at least 10^2 through the use of suitable ion exchange resins on the proper cycle. In time of emergency, ion exchange materials can be utilized in conjunction with standard water treatment procedures to produce water for drinking purposes provided sufficient quantities of resins are available. For example, by placing ion exchange materials on rapid sand filter beds, it is estimated that an overall decontamination factor in the order of 10^3 might be attained. If the 10-day emergency tolerance of 10^{-1} uc/cc is considered, then it would be possible to treat a water containing 100 uc/cc under these conditions. However, the use of ion exchange materials in an emergency could be better adapted to small size units incorporating other decontamination methods such as indicated on page V - 53.

Phosphate Coagulation

Since laboratory findings have indicated that the usual coagulation procedures, in which alum or iron salts are employed with lime have not been too satisfactory in the removal of radioactive contaminants, Lauderdale(13) investigated phosphate coagulation as a means of increasing removals. Such an investigation was considered feasible since many polyvalent cations form relatively insoluable phosphate compounds and the phosphate floc can be formed in a solution of high pH, which favors the removal of many radioisotopes.

Lauderdale investigated individual isotopes by use of jar test procedures similar to those reported for conventional coagulation. The radioisotopes were added to distilled water and a phosphate floc formed by adding either potassium dihydrogen phosphate or tri-sodium phosphate and calcium hydroxide. Maximum removals reported by Lauderdale(13)(14) are shown in Table V-10.

The results indicate that removals of over 99 per cent of Ce^{144}, Zn^{65}, Y^{91}, Nb^{95}, and Zr^{95} can be obtained with phosphate coagulation. Although the conditions of precipitation had little effect on the removal of the above isotopes, marked differences occurred in the removal of Sb^{124} and Sr^{89} when the pH and coagulant dosages were varied. As a result, Sr^{89} was selected for special study. Above a pH of 11.3 and a phosphate to calcium ratio of 2.2:1 (corresponding to a 46 per cent excess of phosphate) there seemed to be little increase in the removal of Sr^{89}.

TABLE V - 10

Removal of Radioisotopes by Phosphate Coagulation(13)(14)

Isotope	Coagulant	Coagulant Dose mg/liter	Removal Per Cent
Ce^{144}	KH_2PO_4	200	99.8
	Na_3PO_4	120	99.9
Sr^{89}	KH_2PO_4	100	81.3
	Na_3PO_4	240	97.8
Y^{91}	KH_2PO_4	100	99.9
Sb^{124}	KH_2PO_4	100	66.1
	Na_3PO_4	120	67.4
Zn^{65}	KH_2PO_4	50	99.6
W^{185}	KH_2PO_4	200	10.7
Zr^{95}	KH_2PO_4	100	99.5
Nb^{95}	KH_2PO_4	100	99.2

Lauderdale carried out coagulation studies using a fission products mixture of the following percentage composition: trivalent rare earths (other than cerium) 43.5, cerium 27.0, strontium 17.4, barium 5.1, ruthenium 2.9, cesium 1.1, other unidentified radioisotopes of low fission yield 3.0. This fission product mixture was diluted with tap water and the initial pH of the waste was raised to 11.5 to 12.0 with NaOH. Removals in excess of 99 per cent (99.2 to 99.6 per cent) were obtained by the addition of 100 ppm Na_3PO_4 and 100 ppm clay or by the addition of 100 ppm $Na_3PO_4$and 100 ppm $Ca(OH)_2$. It should be emphasized that efficiencies of removal will depend on the composition of the waste.

Additional phosphate coagulation studies have been carried out by the RLWRDS, ORNL, using scandium, strontium, tungsten, and yttrium. Radioisotopes of these elements were diluted with tap water, 100 ppm of a local clay turbidity was added, and the initial pH was adjusted to 11.5 by addition of NaOH. A fixed dosage of 3 gpg lime and variable dosages up to 154 gpg Na_3PO_4 were then added. Essentially, the removal results confirm Lauderdale's findings; the maximum removals were: Sr^{89}, 95.5 per cent; W^{185}, 11 per cent; Y^{91}, 99.5 per cent; and Sc^{46}, 99.6 per cent. With scandium, increasing the pH to 11.5 alone effected removals of about 99.2 per cent after settling.

Summarizing, it may be stated that the use of phosphate coagulation for the removal of specific radioelements resulted in removals up to 99.9 per cent. However, these removals are possible only under carefully controlled conditions.

Metallic Dusts

Laboratory jar test studies were carried out by Lacy(7)(15) on the removal of specific radioactive contaminants by various metal dusts. In these experiments tracer quantities of Ru^{106} - Rh^{106}, Y^{91}, P^{32}, Cs^{137} - Ba^{137}, Co^{60}, Zr^{95} - Nb^{95}, $Ce^{141, 144}$ - Pr^{144}, I^{131}, Ba^{140} - La^{140}, Sr^{90} - Y^{90}, MFP - 4 (fission products mixture containing 68 per cent trivalent rare earths and cerium), and MFP - 5 (fission products mixture containing 66 per cent cesium, ruthenium, and rhodium) were added to Oak Ridge tap water. Various amounts of powdered aluminum, copper, iron, or zinc, corresponding to 100, 500, 1,000, and 2,000 ppm of powdered metal, were placed in beakers to which 500 ml of the radioactive solutions were added. After 15, 30, 45, 60, 75, and 90 minutes stirring with the dusts, samples of the supernatant were filtered and counted. In general, it was found that removals increased with increased contact time and dust concentrations.

Removals obtained with 1,000 ppm of the metal dusts at 90 minutes contact are given in Table V-11. In general, highest

TABLE V - 11

Removal of Radioactive Contaminants by Metallic Dusts(15)
(1000 ppm metal dust, 90 minute contact, filtered sample counted)

Contaminant	Stock Solution pH	Initial Activity c/m/ml	Per Cent Removal			
			Fe[a]	Zn	Cu	Al
Ru^{106}-Rh^{106}	8.0-8.2	1,975 - 3,950	99.6	98.4	93.7	92.8
Y^{91}	7.9-8.0	1,610 - 3,230	98.0	98.0	--	--
Zr^{95}-Nb^{95}	7.2-8.3	1,210 - 4,470	99.1	97.9	99.1	99.8
P^{32}	7.8-8.4	128 - 12,000	99.8	98.1	--	84.2
I^{131}	6.5-8.2	4,288 - 6,689	37.2	45.7	42.2	23.2
$Ce^{141,144}$-Pr^{144}	7.9-8.1	3,915 - 4,491	99.9	99.9	99.5	99.8
Ba^{140}-La^{140}	7.9-8.2	4,644 - 10,205	94.8	74.5	65.7	73.8
Co^{60}	8.0-8.2	1,571 - 4,792	92.2[b,c]	--	47.8	30.2[b]
FPM-6	7.2-8.0	4,730 - 10,375	85.8	76.6	92.1	89.4
FPM-5	2.0-7.2	2,890 - 4,220	55.5	39.6	49.3	8.2
Cs^{137}-Ba^{137}	8.0	13,085	8.6	--	--	--

[a] Fe samples were centrifuged before counting
[b] 500 ppm metal dust
[c] 60 minute contact
FPM-6 - Iodine dissolver solution aged 20 days
FPM-5 - Mixed fission product waste containing mainly Cs^{137}-Ba^{137} and Ru^{106}-Rh^{106}.

removals were obtained with iron. Under the conditions reported in Table V - 11, removals with iron were in excess of 90 per cent for the radioisotopes listed with the exception of the fission product mixtures, iodine, and cesium. According to Lacy(15), the primary mechanism involved in the removal of these radiocontaminants is adsorption. Other mechanisms such as metallic displacement, however, may be partially responsible for the removals reported.

Pilot Plant Studies

Coagulation, Sedimentation, and Filtration

At ORNL a 250 gpd (1,000 liter/day) water treatment plant is available for study purposes to help establish basic criteria for building larger pilot plants or full-scale treatment units and to develop optimum operating procedures. The plant has been described in two recent publications(16)(17) and a flow sheet is shown in Figure V - 1. The plant includes facilities for mixing, coagulation or flocculation, sedimentation, and rapid sand filtration.

Preparation of a "raw" water and preliminary mixing of coagulating chemicals is accomplished in two mixing tanks connected in series. Each tank is equipped with a motor driven stirrer. In each tank, the theoretical detention time is 5 minutes at the

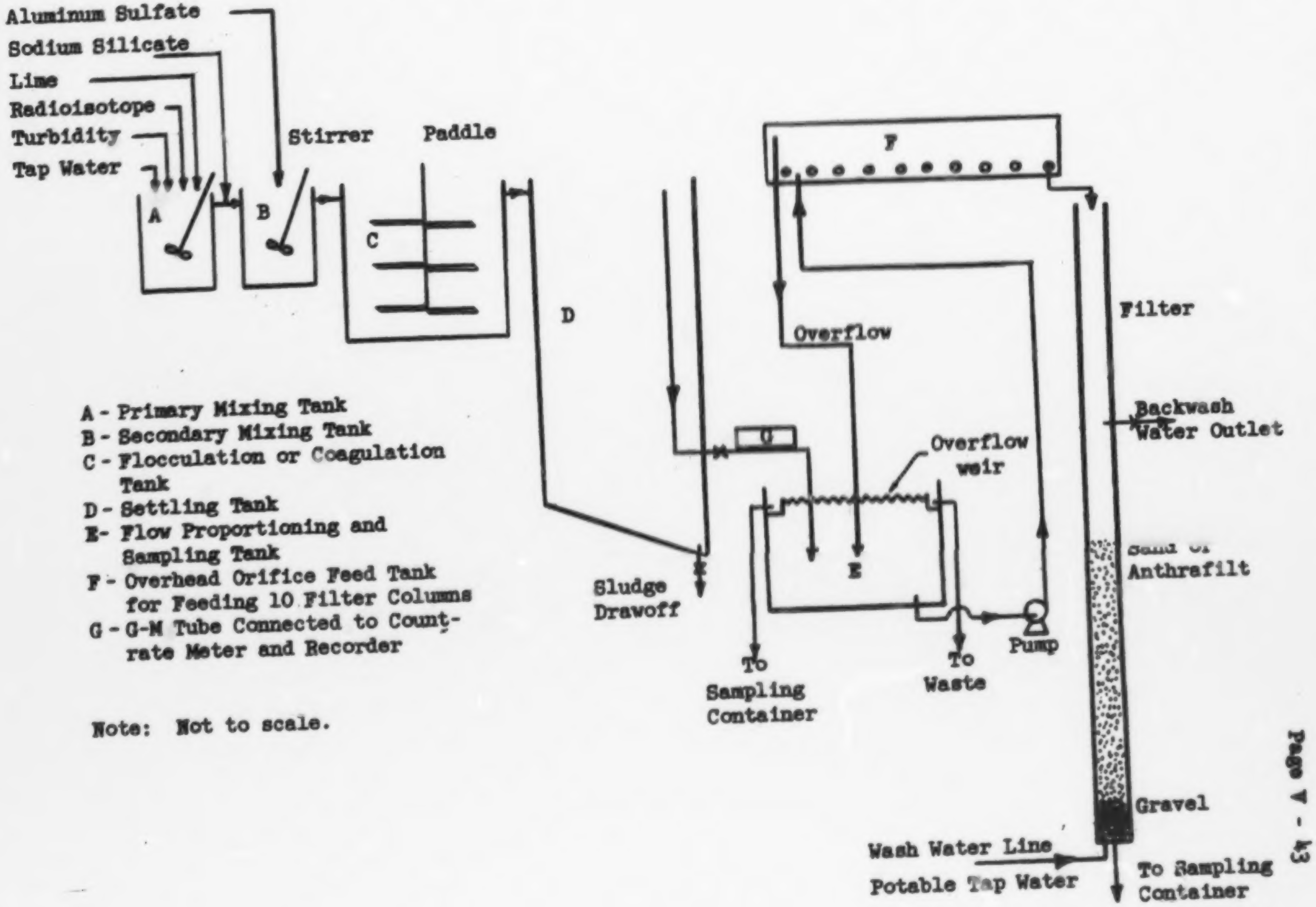
Aluminum Sulfate
Sodium Silicate
Lime
Radioisotope
Turbidity
Tap Water
Stirrer
Paddle
A
B
C
D
E
F
G
Overflow
Overflow weir
Filter
Backwash Water Outlet
Sand or Anthrafilt
Gravel
Sludge Drawoff
To Sampling Container
To Waste
Pump
Wash Water Line
Potable Tap Water
To Sampling Container
A - Primary Mixing Tank
B - Secondary Mixing Tank
C - Flocculation or Coagulation Tank
D - Settling Tank
E - Flow Proportioning and Sampling Tank
F - Overhead Orifice Feed Tank for Feeding 10 Filter Columns
G - G-M Tube Connected to Count-rate Meter and Recorder
Note: Not to scale.

Figure V - 1. FLOW DIAGRAM OF ORNL WATER TREATMENT PILOT PLANT

1,000 liter per day rate; however, this may be varied from 2 to 5 minutes through the use of false bottoms. In recent studies, turbidity and radioactive contaminants together with tap water have been added to the first tank to form a synthetic raw water. Coagulating chemicals, such as lime, sodium silicate, and alum, were fed subsequently -- lime being added to the first tank, sodium silicate added to the discharge pipe from the first tank, and alum added to the second tank.

Descriptions of the other units have been given in the previously mentioned references.

The results obtained in the operation of the pilot plant for the removal of P^{32}, I^{131}, and mixed fission products have been given and discussed in previous publications(1)(16)(17) and will not be repeated in detail here. However, these data are summarized in Tables V - 12 and V - 13.

Pilot plant studies for the removal of P^{31} and P^{32} have been reported also by Eliassen, et al(5) at the Massachusetts Institute of Technology. They measured the removal of P^{31} and P^{32} from Cambridge tap and synthetic Ohio River water. Their findings are summarized in Table V - 14.

In preparing a treated waste solution for further study at ORNL, batch coagulation, sedimentation, and sand filtration were employed for the removal of specific radioisotopes from a fission

TABLE V - 12

Removal of Radioisotopes - Conventional Water Treatment Processes(16)

Radio-isotope	Initial Activity c/m/ml	Turbidity ppm	Alum gpg	Alkali		Sodium Silicate gpg	Carbon ppm	Per Cent Reduction		
				Kind	Dose gpg			Sett.	Filt.	Overall
P^{32}	130-260	95-100	0.52- 0.85	NaOH	0.50- 0.76	--	--	85	79-88	96-98
I^{131}	400-500	95	1.5	Lime	1.5	0.37	--	<0.4	<0.4	<0.4
I^{131}	400-600	100	1.5	Lime	1.5	0.42	4.0	16	11	25

TABLE V - 13

Removal of Mixed Fission Products - Conventional Water Treatment Processes(16) *

Radio-isotope **	Per Cent of Mixture	Turbidity ppm	Dosage			Per Cent Reduction		
			Alum gpg	Lime gpg	Silicate gpg	Settled	Filtered	Overall
Total	100	100	0.9	0.8	0.4	46	44-50	70-73
Ru^{106}-Rh^{106}	15.9					75	0-30	75-82
Ce^{144}-Pr^{144}	12.1					55	46-51	76-78
Y^{91}	17.9					--	40-41	48-50
Sr^{90}-Y^{90}	35.4					61	17-23	68-70
Zr^{95}-Nb^{95}	14.5					68	68-78	90-93
I^{131}	4.2					39	6-53	43-72

* Activity present similar to that in one-month old bomb mixture.

** Ru, Rh, Ce, Pr, Y, and Sr present in chloride form, Zr and Nb as oxalate complex in oxalic acid solution, and I as iodide.

TABLE V - 14

Removal of P^{31}* and P^{32} by a Rapid Sand Filtration Pilot Plant(5)

Run No.	Water	Coagulant	Proportional Removal of P^{31} and P^{32} Over Duration of Run			Decontamination Factor = $\frac{P^{31}\text{ Initial}}{P^{31}\text{ Final}}$
			Coag. + Sett.	Filt. Av. 6 Filt.	Total for Plant	
1	Cambridge	Alum, 100 ppm	74.5	98.30	99.57	232
2	Cambridge	Alum, 100 ppm	81.0	99.62	99.93	1,430
3	Cambridge	$Fe_2(SO_4)_3$, 30-40 ppm	80.9	99.40	99.89	910
4	Ohio R. Turb.=32 ppm.	$Fe_2(SO_4)_3$, 60 ppm	93.7	99.63	99.98	5,000
5	Ohio R. Turb.=32 ppm.	Alum, 48-60 ppm	67.0	98.83	99.61	256

* Initial concentration of P^{31} = 0.01 ppm.

products mixture. The composition of the initial mixture, the various effluents (settled and filtered), and the sludge are indicated in Table V- 15.

From these data it will be noted that even though the composition of this mixture (Table V - 15) is somewhat different from that previously reported (Table V - 13), the removal by the various units comprising the treatment process was practically identical. This is believed to be due to chance inasmuch as removals of individual isotopes in the mixtures varied considerably for the two tests. To determine the removal of specific substances, radiochemical analyses are necessary and should be performed in all instances.

Softening

To confirm his laboratory findings in the removal of Sr^{89} by water softening procedures, Hoyt(9) carried out studies on a pilot plant scale employing the 1,000 gpd Spaulding-type precipitator unit designed by Straub. In this unit, the flow of water is downward through an inverted cone where the added chemicals are mixed, and then upward around the outside of the cone to an outlet weir circling the tank. As the water passes under the cone and upward toward the effluent weir, there is a decrease in velocity due to the increased cross-sectional area through which

TABLE V - 15

Removal of Fission Products Activity by Coagulation, Settling, and Filtration
(Initial concentration of radioactive material - ~ 22,500 c/m/ml)

Radioisotope	Per Cent Composition in				Per Cent Removal		
	Raw	Settled	Filtered	Sludge	Coag.+Settled	Filtered	Overall
Cs	27.2	40.0	17.6	17.2	21.2	75.6	80.8
Sr - Y	27.6	46.5	68.9	20.1	9.5	18.3	26.0
Triv. RE*	31.2	7.9	7.9	53.1	86.3	45.5	92.5
Ru-Rh	3.9	2.4	3.5	4.0	82.1	--	78.7
Ce	8.7	2.9	2.0	9.1	2.1	59.3	92.7
Totals	98.6	99.7**	99.9**	103.5	46.3	44.4	70.1

* Trivalent rare earths other than Ce
** Quantitative analytical recoveries corrected to approximately 100 per cent.

the liquid passes. As a result the floc particles eventually reach a state of equilibrium, at which they are held by the upward velocity of the water and the downward gravitational force.

The results of the pilot plant investigation are listed in Table V - 16 along with the results of jar test studies. These data indicate that higher removals were obtained in the pilot plant than in jar tests under similar levels of chemical dosage. The removal of all suspended matter from the precipitator effluent (centrifuging or sand filtration) increased the removals of hardness and activity to 87.3 and 96.0 per cent, respectively, when 50 ppm excess lime-soda ash was added.

Emergency-Type Water Treatment Units

If a source of water supply suffers a direct hit, the concentration of radioactive materials resulting from the blast may be sufficiently high so as to preclude the production of a safe water supply by the usual water treatment processes. Following such a catastrophe, all householders should be warned immediately that, until further notice, the water supply is unsafe for human consumption. They should be advised that they have adequate water for their most immediate drinking and culinary needs in their hot water tanks, or if these are not available, they should resort to the use of canned fruit juices, soft

TABLE V - 16

Pilot Plant and Jar Test Results - Softening Experiments(9)

Treatment	Removal of Activity - Per Cent		Removal of Hardness - Per Cent	
	By Jar Test	By Spaulding Precipitator	By Jar Test	By Spaulding Precipitator
Required Doses	75.9	79.5-86.0	52.5	59.1-68.1
Plus 50 ppm Excess*	80.1	86.8	56.2	76.0
Plus 100 ppm Excess*			67.3	85.5

* Lime-soda ash excess.

drinks, etc., until emergency water supply points have been established. If the water is grossly contaminated with radioactive materials, emergency supplies may be procured from existing safe ground water supplies, they may be brought in from other areas by tank truck or tank car, or use may be made of field decontamination units. Where evaporation facilities are available in certain industrial establishments that have not been damaged, these also may be employed to produce sufficient safe water to meet minimal requirements.

Tests have been made on the removal of radioactive contaminants by several units that have been proposed for treating relatively small quantities of water under emergency conditions. Most of the units of this type studied to date have had as their primary mechanism of removal either ion exchange or adsorption. The results obtained are indicated below along with a description of the unit and other pertinent data.

Filtr-Ion Unit

The Filtr-Ion Unit, a product of the LaMotte Chemical Company, is an ion exchange unit which contains Amberlite "monobed" resin MB-3 (IR-120 and IRA-410), and is designed to produce chemically pure water from tap water. This unit is reportedly capable of producing 10 gallons of water equivalent in quality to triple-distilled water at a cost of $3.85 per unit. Exhaustion of the resins (from the hardness standpoint) is indicated by a color change from black to yellow.

In an evaluation of the unit(18), 34 liters of solution containing 94,300 c/m/ml of fission products activity was passed through the resin. The first 500 ml of effluent counted about 10 c/m/ml and the 34th liter, 2,750 c/m/ml, for removals of 99.99 and 97.1 per cent, respectively.

Multi-Bed Decontamination Unit

One of the more promising units for the emergency treatment of small volumes of radioactively contaminated liquids was that developed by R. A. Lauderdale and A. H. Emmons, ORNL(19)(20). In their unit, radioactively contaminated solutions were passed through layers of steel wool, burnt clay, activated carbon, and mixed cation and anion exchange resins. The initial concentration of 2.5 uc/cc was reduced to less than 10^{-4} uc/cc for a removal in excess of 99.996 per cent. The unit is capable of treating approximately 30 liters of water before significant breakthrough of radioactive materials occurs.

Radiochemical analyses of activity adsorbed by the materials in the decontamination unit are reported in Table V - 17.

TABLE V - 17

Removal of Radioactive Contaminants - Lauderdale and Emmons Unit(19)(20)

Isotope	Per Cent of Total Removal				
	Steel Wool	Burnt Clay	Activated Charcoal	Cation Resin	Anion Resin
β	85.26	11.99	0.80	1.92	0.21
Ru	95.96	3.62	0.14	0.19	0.09
Zr	99.39	0.43	0.061	0.108	0.018
Sr	69.72	18.27	3.56	8.37	0.81
RE*	86.81	13.02	0.09	0.06	0.003
Ce**	97.13	2.84	0.003	0.02	0.002
Cs	59.74	39.99	--	0.14	0.13
Nb	98.7	0.58	0.12	0.44	0.16
Te	98.34	0.89	0.19	0.237	0.331

* Total rare earths.

** Cerium is also included with the total rare earths.

Field Emergency Units

Felton R. Nease(21) reported some preliminary results on the passage of strontium and cesium through beds of leaves and clay. The materials used were from the White Oak Lake region and were as nearly uncontaminated by radioactive materials as possible. As a result of his studies, he concluded that dead organic plant matter accumulated more radioactive material than did similar living organic matter (see also Page IV - 9). He found that about 53 per cent of the strontium and cesium was removed on the first pass and that the overall removal was increased to 9[illegible].7 per cent following the second pass through the improvised filter.

Following the initial experiments reported by Nease, further investigation of this method of decontamination was made by Lacy(7). His unit consisted of a column containing several 5 cm layers of sand, gravel, humus, coarse vegetation, and clay. A total of 17.8 liters of a solution containing a mixture of radioisotopes was passed through the unit for an overall removal (filtered effluent) of 95.1 per cent. The removal amounted to 94.4 per cent when the effluent was unfiltered, that is, as discharged from the decontamination unit.

Field Distillation Units

Another type of field unit that may be utilized in areas where the water supply has been grossly contaminated with radioactive material and where no other source of water is readily available is the field distillation unit similar to the type used by the military forces. Various types of evaporators are used to concentrate high level radioactive wastes at several AEC installations. Decontamination factors of 10^5 or more have been reported for such units in conjunction with various entrainment devices. Similar distillation of contaminated wastes may produce an effluent suitable for drinking purposes.

Summary of Water Decontamination Results

Generalizations regarding efficiencies in removing radioactive contaminants from water by commonly utilized water treatment procedures are difficult to formulate. As emphasized throughout this report, the extent of decontamination which treatment processes provide is largely dependent on test conditions and the chemical composition of the water involved; however, it is possible to specify optimum treatment procedures for individual isotopes. In many instances, the effects of pertinent variables on removal efficiency have been studied and the decontamination range which might be anticipated in actual practice can be estimated with fair accuracy.

The optimum removals which can be expected for specific isotopes utilizing the various types of treatment processes thus far investigated are summarized in Table V - 18. These estimates of removal efficiency represent a composite of all information reported herein. An operating procedure for each of the processes (noted in Table V - 18) was selected which would give maximum removal efficiencies and yet be within practicable limitations. With softening, for example, higher dosages of lime and soda ash (>10 gpg) would increase removals somewhat for certain isotopes, but such higher dosages probably would not be justified economically. With coagulation, it has been arbitrarily assumed that a moderately turbid water would be treated, i. e., turbidity in the order of 100 ppm.

The elements investigated have been arranged in accordance with Periodic groupings in Table V - 18. In the two groups where comparisons can be made (Groups II and III), the data appear fairly similar for elements in the same grouping. This agreement may or may not be significant, and it should not be implied that all elements in the same group will react similarly even though the conditions of treatment may be identical.

With wastes containing mixtures of radioisotopes, such as the more common mixed fission product type wastes, a rough estimate of removal efficiency could be obtained for a given

TABLE V - 18

Optimum Removals of Radioisotopes from Water

(Initial activities varied from 200 to 20000 c/m/ml)

Treatment Process	Removal of Radioisotopes - Per Cent*															
	Group I	Group II				Group III		Group IV,V			Group VI		Group VII	Group VIII		Rare Earths
	Cs	Zn	Sr	Cd	Ba-La	Sc	Y	Zr-Nb	P	Sb	Cr	W	I	Co	Ru	Ce-Pr
Columns																
Cation exchange	99+		99+	95-99	95-99	95-99	75-90	25-75				0-25				
Anion exchange	0-25		0-25	0-25	25-75	95-99	95-99	99+				95-99	99+			
Mixed resins	99+		99+	99+	99+	95-99	95-99	99+	99+			95-99	99+			99+
Greensand			99+		95-99	95-99	25-75					0-25				
Filter sand	0-25		0-25	0-25	0-25	90-95	75-90	75-90				0-25				
Turbidity																
100 ppm	25-75		0-25	0-25	25-75	25-75	25-75	90-95				0-25				25-75
5000 ppm	95-99		25-75	25-75	75-90	95-99	90-95	95-99				25-75				
Softening																
10 gpg lime) 10 gpg soda ash)	0-25		90-95	99+	90-95	90-95	90-95	75-90				0-25				
Phosphate Coagulation-100 ppm $PO_4^{\equiv}$		99+	75-90			99+	99+	99+		25-75		0-25				99+
Coagulation																
6 gpg $FeSO_4$, $FeCl_3$ or alum	25-75		25-75	75-90	25-75	95-99	95-99	95-99	95-99		95-99	75-90	0-25	0-25		90-95
Metallic Dust																
1000 ppm Fe	0-25				90-95		95-99	99+	99+				25-75	90-95	95-99	99+

* Removals are indicated by the following class intervals: 0 to 25, 25 to 75, 75 to 90, 90 to 95, 95 to 99, and 99+. These are not ranges of removals. It merely indicates that the removal value fell within the indicated interval.

treatment process if the radiochemical content of the waste were known and removal values were available for the individual components. In any case, however, a far more precise estimate would be obtained if the waste in question is treated on a laboratory scale.

Very little is known, at this time, as to the exact mechanisms of removal of radioisotopes by water treatment processes. This is primarily due to the extremely low concentrations (10^{-7} to 10^{-10} ppm) of contaminants encountered in these studies. The degree, as well as the mechanism, of removal may also vary considerably for waters of differing composition. Most of these studies have been conducted using Oak Ridge tap waters contaminated with rather pure solutions of radioisotopes, and with few exceptions, removals from other potable waters or waters containing complexing chemicals such as detergents have not been investigated.

REFERENCES

1. Straub, Conrad P., Morton, R. J., and Piacak, O. R. Studies on the Removal of Radioactive Contaminants from Water. J., American Water Works Assn., 43: 10, 773 (October, 1951).

2. Lauderdale, R. A. Unpublished material, Oak Ridge National Laboratory, Oak Ridge, Tennessee (1950).

3. Straub, Conrad P., Brockett, T. W., Jr., and Stepp, Robert. Preliminary Reports of Progress - Laboratory Studies. Water Decontamination. III. Studies with Ce^{144}, Y^{91}, and I^{131}. Memorandum Report, Oak Ridge National Laboratory, Oak Ridge, Tennessee. Central Files Number 51-5-202 (May 3, 1951).

4. Straub, Conrad P. and Krieger, Herman L. Removal of Radioisotopes from Waste Solutions by Soils. Soil Suspension Studies. Presented at the 8th Industrial Wastes Conference, Purdue University, Lafayette, Indiana, May 6, 1953.

5. Eliassen, Rolf, Kaufman, Warren J., Nesbitt, John B., and Goldman, Morton I. Studies on Radioisotope Removal by Water Treatment Processes. Journal, American Water Works Assn., 43: 8, 615 (August, 1951).

6. Swope, H. Gladys. Discussion of Eliassen, *et al*. paper (see Reference 5). Journal, American Water Works Assn., 43: 8, 633 (August, 1951).

7. Lacy, William J. Unpublished material, Oak Ridge National Laboratory, Oak Ridge, Tennessee. (1952).

8. Christenson, C. W., Ettinger, M. B., Robeck, Gordon G., Hermann, E. R., Kohr, K. C., and Newell, J. F. Removal of Plutonium from Laboratory Wastes, Ind. Engr. Chem., 43: 7, 1509 (July, 1951).

9. Hoyt, W. A. The Effect of the Lime-Soda Water Softening Process on the Removal of Radioactive Strontium. Thesis submitted to North Carolina State College for Master's Degree (1952).

10. Gloyna, E. F. Some Basic Investigations in the Treatment of Water Contaminated with Fission Products. Unpublished material, Oak Ridge National Laboratory, Oak Ridge, Tennessee (1950).

11. Ayres, John A. Treatment of Radioactive Waste by Ion Exchange. Ind. Engr. Chem., 43: 7, 1526 (July, 1951).

12. Friend, A. G. Report on the Investigation of the Removal of Iondine (I^{131}) and Strontium (Sr^{89}) from Water by Ion Exchange Resins. Thesis submitted to Virginia Polytecknic Institute, Blacksburg, Virginia (June 8, 1952).

13. Lauderdale, R. A. Treatment of Radioactive Water by Phosphate Precipitation. Ind. Engr. Chem., 43: 7, 1538 (July, 1951).

14. Lauderdale, R. A., Jr. Studies on the Removal of Radioisotopes from Liquid Waste by Coagulation. Oak Ridge National Laboratory, Oak Ridge, Tennessee. ORNL-932 (January 23, 1951).

15. Lacy, W. J. Removal of Radioactive Material from Water by Slurrying with Powdered Metal. Journal, American Water Works Assn., 44: 9, 824 (September, 1952).

16. Straub, Conrad P., Morton, Roy J., and Placak, Oliver R. Oak Ridge Reports Results on Water Decontamination Study. Engr. News-Rec., 147: 7, 38 (August 16, 1951).

17. Straub, Conrad P. Removal of Radioactive Waste from Water. Nucleonics, 10: 1, 40 (January, 1952).

18. Lindsten, Don C. Unpublished material, Oak Ridge National Laboratory, Oak Ridge, Tennessee. (1952).

19. Lauderdale, R. A. and Emmons, A. H. A Method for Decontaminating Small Volumes of Radioactive Water. Journal, American Water Works Assn., 43: 5, 327 (May, 1951).

20. Lauderdale, R. A. and Emmons, A. H. Decontamination of Small Volumes of Radioactive Water. Nucleonics, 8: 5, 21 (May, 1951).

21. Nease, Felton R. Unpublished material based on data gathered by TVA under AEC sponsorship. Oak Ridge National Laboratory, Oak Ridge, Tennessee. (1951).

SECTION VI

INSTRUMENTATION

Satisfactory commercial counting instruments are available for measuring the concentrations of radioactive materials usually encountered in practice or those employed in the routing laboratory investigations reportes in Section V. Since these instruments are readily available they will not be described herein.

The problem of measuring the amount of decontamination that has taken place as a result of the employment of a specific waste treatment or decontamination process is only one of the many problems that one is faced with in measuring the concentration of radioactive materials in a given environment. It has been necessary, therefore, to develop special types of instrumentation where these are not available commercially, or where the available types proved inadequate, for the measurement of the concentrations of radioactivity in rain, ground, and surface waters, in waste streams, river muds, and other deposits, and in biological life.

Because of the wide variety of uses, as indicated above, for which instrumentation is required, it has been impossible to develop a single type of effective instrument. Rather, it has been necessary to develop specialized equipment to meet specific needs. The information reported herein as to sensitivity and suitability must be interpreted with these conditions in mind.

Column Type Monitor

An ion exchange column type water monitor(1) has been developed for the detection of low levels of radioactivity in liquids, and consists of a 10-inch, thin-walled (30 mg/cm^2) glass G-M tube surrounded by a bed of Dowex-50 cation exchange resin. In operation the resin concentrates the radioactive material (cation plus some particulate matter) from the volume of liquid passing through the bed. The buildup in activity is recorded on a strip chart as a line of constant slope for each concentration of activity. The slope of this line, therefore, may be used as a measure of the concentration of radioactive materials present in the liquid. Calibration of the instrument is accomplished by the use of tap water spiked with known amounts of appropriate radioactive materials.

This monitoring device has been used to routinely measure the concentration of radioactive materials in rain water samples and to a lesser extent to measure the amount of radioactivity in ground and surface waters in the Oak Ridge area. It has measured cationic activities in the order of 10^{-6} to 10^{-7} μc/cc in rain water samples. Instrument sensitivity decreases with increasing salt concentration (less water passed through the unit); and varies with the particular radioisotope involved, its chemical characteristics, concentration, energy spectrum, type of radiation, etc. Accordingly, sensitivity cannot be generally defined but must be determined by calibration under conditions similar to those employed under test.

Aside from the relatively high sensitivity of this device, another important advantage as compared with other immersion-type units, is that the G-M tube is decontaminated during regeneration of the resin column. Therefore, buildup of activity on the tube itself should present no problem. Presumably, this instrument could be adapted to continuous monitoring by employing two columns with the proper control equipment.

Centrifugal Type Monitor

Two versions of an instrument for monitoring liquids, which use the principle of centrifugal force, have been developed and have been reported in an ORNL report(2).

One of these is a continuous flow water-monitoring cell into which the water is injected tangentially within a confining cylinder. The injected stream becomes a rotating cylindrical wall of water with a hollow center. When equilibrium is established between the inflow and the outflow, the G-M tube detector is lowered into the hollow center after the water wall is established. Regulation of the water flow does not appear to be too critical. The only beta absorber between the water wall and the detector is a 3/16" annular air gap and the wall of the detector.

A variation of the above instrument was developed for the purpose of calibrating this continuous flow monitor. It consists essentially of a flanged polyethylene sample cell of 131 ml total capacity placed in a hollow rotor fixed to a 1800 rpm motor shaft. When 80 ml of sample is placed in the cell and the cell is rotated, centrifugal force forms a vertical water wall leaving an air space into which the detector is lowered. When solutions containing alpha radiation or low energy betas are to be measured, a cell of smaller volume is used. This instrument also provides a very useful and rapid method of measuring radioactivity in liquids of 15 ml to 80 ml volume.

The choice of a radiation detector is influenced by the energy spectrum of the radioisotopes present. Two types of detectors have been used. When the activity consists of mixed fission products or of beta emitters of about 0.7 Mev or higher energy the detector suggested is a thin wall (30 mg/cm^2) G-M tube such as the RCL, Mark 1, Model 11. When the problem is the detection of beta particles of less than 0.7 Mev energy or of alpha radiation, an alpha-beta detector sensitive to incident betas of about 20 Kev or higher has been designed. This consists of a cylindrical G-M counter with walls of rubber hydrochloride (0.7 mg/cm^2). For electrical conductivity the inner surface of the shell is coated with a film (50 $microgram/cm^2$) of gold. The shell is rugged and may be collapsed without injury. In operation the cell is flushed continuously at a flow rate of about 10 cc/min with a gas mixture containing 10 per cent methane and 90 per cent argon.

With the most sensitive detector, relatively low levels of activity may be measured. Potassium-40 naturally present in body fluids is detected easily, Sr^{89} activity in water may be detected at a concentration of 1×10^{-6} μc/cc, and Tl^{204} at a concentration of 3×10^{-6} μc/cc. Alpha radiation (from Pu^{239}) may be measured at

4×10^{-5} μc/cc. A partial evaporation will permit better detectability at lower concentrations; e.g., reduction in volume of the sample from 800 to 80 ml, will lower the concentration which can be detected by a factor of 10.

For many uses, the advantages of this monitoring instrument are that no evaporation or drying of the sample prior to counting is required, low energy radioisotopes are detectable, and contamination of the tube does not result as in the case of immersion detectors. As an example, operation for one hour with a solution of Sr^{90} - Y^{90} activity at a concentration of 10^{-1} μc/cc resulted in no measurable contamination of the counter tube.

Other Types of Continuous Monitors

Several other systems for continuously monitoring water have been devised and are of interest. In one of these(3), three mica end-window G-M counters are positioned immediately above a tray containing running water. The three counters have a combined background counting rate of about 60 c/m (shielded) and a total beta window area of about 3 square inches. A counting rate circuit records continuously on a strip chart.

In another system for continuous monitoring(4), the radiation detector is a glass G-M counter tube at the center of a circle of smooth-flowing individual vertical water streams. The counting rate is recorded on a strip chart.

At least two continuous water monitoring systems are available commercially. In each of these systems, the detecting device is immersed in the water and this may cause an apparent increase in the activity measurements due to accumulation of radioactive materials by biological life adhering to the surface of the tube. Another disadvantage of these commercial systems is that the detecting devices must be calibrated in situ so that counting geometry will remain unchanged. The sensitivity level of these two systems is in the range of the emergency permissible concentrations, i.e., $\sim 10^{-2}$ μc/cc.

In one of these commercially available monitors(5), the water is passed through a jacketed thin wall G-M tube and the circuitry is such that the resulting counting rates are recorded on a chart. Alpha particles must be counted separately on dried samples.

The second of these commercially available continuous monitors(6) has available four different systems of varying degrees of accuracy and with various methods of recording data. All of them employ large stainless steel beta-gamma G-M tubes which are mounted directly in the water pipe. The tube is completely waterproof. These systems may also use a scintillation detector which is sensitive only to alpha radiation and which must not come into direct contact with the water. Where the activity is to be continuously monitored, the alpha detector is mounted over a moving belt of filter paper which is presumed to pick up particles from the water and bring them under the detector.

Monitoring Systems

Monitoring devices are sometimes required at inaccessible or distant locations and it becomes necessary to provide instantaneous measurements or radioactivity at a more convenient station. In other instances, a need has arisen for monitoring several points in a flow system where a minimum amount of recording equipment is desired. To exemplify monitoring systems of these types, the waste monitoring system at ORNL and a four-point monitoring system are described.

Waste Monitoring System at ORNL

In addition to the intermediate monitoring which is the responsibility of the Operations Division, the heterogeneous radioactive process wastes are also monitored by the RLWRDS at two points before discharge into the Clinch River. The semi-final monitoring step occurs at the outlet end of a settling basin where the installation consists of a weir box for flow measurement and sampling of the waste stream. Included are a standard 90° V-notch weir, a USGS-type float well and water level recorder, a Trebler sampler, and space for continuous monitoring instruments.

The final monitoring step occurs at the dam at the outlet of White Oak Lake. Because of the necessity of providing a system that would furnish accurate, reliable, and continuous records of the amounts of water and of gamma radioactivity discharged from the Lake, considerable development work was required. Since the point of discharge is rather isolated, it was also desirable that these measurements should be telemetered to the Laboratory so that any irregularities would be detected instantly. Such a system is flexible and has been developed with an eye toward future needs and to permit the incorporation of improvements in instruments and equipment.

The system has three main components (1) a water level recorder, (2) a radiation detection assembly, and (3) a telemetering system. Water level measurements are made with a standard USGS-type float well and water level recorder. The remainder of the system was developed as a part of this program and is described in the following paragraphs.

In the radiation detection system, a boom assembly is used to hold a radiation detector just over the water surface. The distance between the detector and the water surface is automatically controlled and held constant by means of a servo-mechanism or it may be manually controlled. A safety device is also incorporated so that, in event of any failure - electrical, mechanical, or manual - which might occur, the detector will never become contaminated due to immersion in the Lake. The detector is a scintillation counter composed of an anthracene crystal, photomultiplier tube, pre-amplifier, and a cathode follower, all encased in a lead cylinder two inches thick and having a thick window on the bottom. The geometry is such that the detector is unidirectional. Because of the thick window, the detector counts only gamma radiation, these counts being fed into a specially designed count rate meter. Care has been exercised in the design and development of these instruments so that they would not be sensitive to weather variations. Work is in progress to incorporate a beta detector into the system.

The telemetering system (carrier current type) and associated equipment provide an accurate and continuous record of water and radioactivity levels at a convenient location in the Laboratory. An alarm attracts attention at any time when either of these levels exceed certain predetermined values. Auxiliary standby equipment is stationed at all points in the system and is immediately available for use should there be any failure.

Multiple Point Monitoring System

To meet the requirements of minimum equipment at minimum expense, a multiple point monitoring system was developed which permits the continuous semiquantitative measurement of the concentration of radioactive materials at several points. As developed for four point monitoring, the equipment consists of four radiation detectors connected by coaxial cables to a switching unit, a count rate meter with high voltage supply, and a recorder. A manual or automatic switching unit provides separate high voltage adjustment for each detector. When automatic switching is used, a timing motor drives a rotary switch that permits the signal from each detector to reach the count rate meter in a prearranged sequence. In practice, the solution to be monitored was passed through Tygon tubing wrapped around the sensitive portion of the G-M detector tube.

River and Lake Survey Equipment

After wastes have been monitored and discharged, it is desirable to survey the resulting situation in receiving bodies of water. This may be done accurately, but laboriously, by the collection and analysis of large numbers of samples of water, muds, and other materials. This method is used but instruments for more rapid surveys have also been developed.

Two instruments initially developed for survey purposes, although partially successful, were discarded for various reasons; however, these instruments are described briefly. The first instrument developed by the Section for use in water and mud surveys consisted of copper-walled G-M tubes and a cathode follower circuit housed in a large galvanized can. This was connected to a portable count rate meter by means of an electrical cable and lowered into the water by a rope. The principal disadvantages of this instrument were its bulkiness, awkwardness in handling, and the fact that it was not designed to withstand pressures at great depths. The objective of the second survey instrument, a "trolling probe", was to estimate the relative amounts of radioactivity in various sections of a body of water by towing the instrument behind a boat. The instrument consisted of a special brass-walled G-M tube, one inch outside diameter and 3 feet long, with a waterproofed connection to a short length of coaxial cable. Pulses from this probe were fed through the cable to a survey meter adapted for this purpose. Use of this device was discontinued because another instrument (called the "flounder") was developed which proved more convenient and sensitive for the same purpose and could also be used for measuring the accumulation of radioactivity in the bottom muds. Actually, two similar instruments are used, one for beta and the other for gamma readings, differing only in external housing.

These instruments consist of 12 thin-wall G-M tubes and a cathode follower encased in a waterproof housing (15 1/2" x 15 1/2" x 1 1/2"). For the gamma instrument two sides of the housing are made of 1/4" lucite and in the other the sides are made of 1/32" cellulose acetate. The "flounders" are connected to count rate meters (or scalers) and power supplies by 72 feet of rubber covered cable. They can be trolled in water behind a boat, lowered to various depths up to 70 feet for detection of radioactivity, or lowered to the bottom of a body of water for measurement of radioactivity in the mud. The "flounder" was primarily adapted for gamma-ray measurement in terms of mr/hr. To date this instrument has been used for relative activity measurements of water and bottom deposits. For more precise work, calibration in terms of μc/cc of water or μc/gram of mud for the radioelements to be expected in a given location would have to be made.

Survey measurements of the activity in river bottom sediments may be made from a boat by lowering the "flounder" to the river bottom. For example, measurements made along the Tennessee River upstream from Oak Ridge, generally fell within the normal range of background. Slight increases were noted in the Tennessee River below its confluence with the Clinch River. These increases disappear directly below each of the dams because of the scouring action on the river bottom caused by increased water velocity.

Well-Logging Equipment

The measurement of levels of radioactivity in surface waters or associated components of these systems such as bottom sediments is only part of the general problem. In any area that includes settling basins, burial grounds, or similar installations, an evaluation of the ground water is desirable. In the X-10 area, fifty-five wells have been drilled in selected locations for monitoring purposes. These wells may be sampled for chemical and/or radiochemical analysis and counting purposes, and essential information is obtained in this manner. This method, however, is too laborious for routine monitoring. To facilitate these measurements, well-logging equipment has been developed. This equipment consists of a thin-walled aluminum probe containing G-M tubes, preamplifier, and impedence matching network connected to about 310 feet of cable to a counting rate meter and an Esterline-Angus recorder. The probe is lowered into the well at a predetermined uniform rate by a motor driven cable reel. Since the unit is equipped with an auotmatic cut-off, the constant attention of an operator is not required. Activity versus depth is recorded on a strip chart.

Cosmic radiation, natural radioactivity of the geological formation, and the ratioactivity of ground waters contribute to the total measured count. No attempt has been made as yet to correct exactly for cosmic radiation and radiation from stratigraphic layers. The instrument has proved very successful, however, for relative measurement of radioactive materials in ground waters. In this way, changes in activity can be quickly and conveniently noted, and, if necessary, samples of the water can be accurately counted and radiochemical analyses made. To determine the exact sensitivity of the probe, calibration for specific type waters and radioisotopes would be required.

Miscellaneous Measurements of Radioactivity

In addition to monitoring wastes and surveying the receiving body of surface water, other survey measurements are desirable. Some of the instruments devised for making these measurements will be described briefly.

A mud probe has been devised to estimate the radioactivity of muds at depths from one to eighteen inches below the surface. The instrument consists of a thin-wall stainless steel G-M tube detector 3/8 inches outside diameter, 6 inches long, with a sensitive length of one inch near the tip. The detector is mounted on a pointed aluminum rod and is extendable from three feet to 13 feet enabling use in water at depths up to 12 feet. The detector is pushed into the mud to various depths and readings observed on a Tracerlab Survey meter which has been modified for use with the probe. The instrument has been calibrated in mr/hr using a radium standard and has a range of 0.01 to 100 mr/hr on four scales. A beta detector has been designed, but has not been constructed as yet.

In another of our investigations, it became necessary to estimate the amount of activity in fish without sacrificing the fish. This measurement was facilitated by designing a special watertight probe consisting of a handle, a 3/8 inch outside diameter stainless steel G-M tube, and about four feet of cable. Initially, two types were constructed: (1) gamma probes with G-M tubes of thick stainless steel, and (2) beta probes with G-M tubes having thin ribbed stainless steel walls (0.0018 inches ± 0.0005 inches). These probes are used with a battery operated scaler. The G-M tube is inserted in the mouth of the fish for making measurements.

Field tests suggested several improvements which have been incorporated into later gamma probes. Improved probes have been constructed using copper G-M tubes lined with bismuth instead of stainless steel tubes. A scintillation type gamma detector using sodium iodide crystals has also been designed.

Calibration of the probes is accomplished by taking readings on a number of radioactive fish and then sacrificing the fish for assay.

Emergency Water Monitoring

Commercially available instruments have been evaluated for emergency monitoring. Hursh, Zizzo, and Dahl(7) have reported some work utilizing survey meters for water monitoring. Additional studies have been carried out by Gerstein and reported in the AWWA Task Group Report(8). A rather comprehensive evaluation(9)(10) of representative types of commercially available instruments has also been carried out as a part of this program and will be reviewed briefly.

In studies with G-M survey meters(9) three representative types of instruments with five interchangeable probes were used. Readings were made by holding the probe directly over the surface of the liquid and also by immersing the probe into the liquid. To protect the probe against contamination, it was encased in a thin rubber sheath. Solutions of various individual radioisotopes and of mixed fission products were used for test purposes and for obtaining calibration curves. A sample of unknown concentration is monitored in terms of milliroentgens per hour, and these readings are converted to concentrations in mircocuries per milliliter by means of calibration curves. It was concluded that fairly accurate measurements could be made for activities in the range of 10^{-1} to 10^{-4} uc/cc provided the survey meters were calibrated using samples of similar radiochemical composition and using the same conditions of geometry.

Evaluations of two types of electroscopes were carried out under four operating conditions, i.e., four methods of sample preparation(10). A number of radioisotopes having different beta and gamma energies were used and it was found that the response of the instrument varies with the type and energy of radiation. The activity range which may be measured by use of the Landsverk electroscope (Model L-75) is about 10^{-6} to 10^{-2} uc/cc for alpha particles and about 10^{-4} to 1 uc/cc for beta particles.

Comparison of the operating characteristics of G-M survey meters and electroscopes in measuring activity in water samples indicates that both are sensitive to activity concentrations in the emergency tolerance range ($\sim 10^{-2}$ uc/cc). The electroscope offers some advantage in that it is sensitive to alpha radiation as well as beta radiation. Both types of instruments require calibration with standards of approximately the same energy composition as the material under study. With mixed fission products, thallium-204 standards could be prepared and distributed for calibration. Either type of instrument should be readily available in time of emergency although the ruggedness, simplicity, and easy maintenance of the electroscope offer certain advantages over the G-M survey meter.

Summary

Selection of a method for detection and measurement of radioactivity in water is primarily dependent upon the particular purpose involved. All detection instruments are, to a certain extent, dependent upon the energy and type of radiation released. In measuring liquids of unknown radiochemical composition, a relative, rather than an absolute, activity concentration is obtained. For many purposes, relative measurements are adequate to denote trends or changes in activity content. Where absolute measurements are required, careful calibration of the detecting device is necessary.

Of the instruments discussed in this section, the column and centrifugal types show promise in the continuous monitoring of liquids of low level activity (10^{-4} to 10^{-7} uc/cc). As yet these instruments have not had widespread use, however. Commercial instruments, of the immersion type are suitable for measurements of activity in the order of 10^{-1} to 10^{-3} uc/cc provided contamination of the detecting device presents no problem. Other instruments have been developed for river and lake surveys including monitoring of bottom sediments, for well-logging, and for monitoring activity in biological samples such as fish. With regard to emergency monitoring of water supplies, the electroscope and G-M survey meter have been evaluated and both appear adequate in the event of serious water contamination.

REFERENCES

1. Emmons, A. H. and Lauderdale, R. A. A Method for Estimating Low Levels of Radioactive Contaminants in Water. Nucleonics, 10: 6, 22 (June, 1952).

2. Hurst, W. M. Monitoring of Liquids for Radioactivity. Oak Ridge National Laboratory, Oak Ridge, Tennessee. ORNL - 1155 (February 26, 1952).

3. K-837.

4. HW-9584.

5. Radiation Counter Laboratories, Inc. Leaflet entitled "Radiological Defense Protective Devices."

6. Tracerlab, Inc. Booklet "Radiation Monitoring for Civil and Industrial Safety."

7. Hursh, J. B., Zizzo, S., and Dahl, A. H. Survey Meters for Water Monitoring. Nucleonics, 9: 5, C-16 (November, 1951).

8. AWWA Task Group Report. Instrumentation and Methods for Testing Radioactive Contamination in Water, Journal, American Water Works Assn., 44: 7, 583 (July, 1952).

9. Lacy, W. J. Use of G-M Survey Meters for Radioactive Water Monitoring. Oak Ridge National Laboratory, Oak Ridge, Tennessee. (1952).

10. Kahn, B. Use of Electroscopes for Radioactive Water Monitoring. Oak Ridge National Laboratory, Oak Ridge, Tennessee. (1952).

May 18, 1953

VI-19

APPENDIX A

Staff Members
Radioactive Liquid Waste Research and Development Section

ORNL

Roy J. Morton, Leader
T. W. Brockett, Chemist
M. J. Cook, Biologist (part time)
J. M. Garner, Physicist
B. Kahn, Chemical Engineer
B. Reed, Secretary
H. J. Wyrick, Phy.Sci.Aide
*A. H. Emmons, Chemist
*V. I. Knobf, Health Physicist
*R. A. Lauderdale, Chemical Engr.
*K. S. Lyle, Phy. Sci.Aide
*R. M. Nichols, Phy. Sci.Aide
*R. Stepp. Phy. Sci. Aide

USPHS

O. R. Placak, Sr. Scient., Asst. Leader
C. P. Straub, Sr. San. Engr.
D. A. Pecsok, Sr. Asst. San. Engr.
M. S. Seal, Sr. Asst. San.
M. W. Carter, Sr. Asst. San. Engr.
H. L. Krieger, Chemist
F. Kalil, Physicist
*E. C. Tabor, Chemist

TVA

**L. A. Krumholz
**E. R. Eastwood
**W. T. Helm
**W. T. Miller
*L. R. Setter
*O. W. Kochtitzky
*F. R. Nease
*R. Ross
*D. W. Yambert

U. S. Army

W. J. Lacy, ERDL, Chemist
F. L. Cobler, ERDL, Phy.Sci.Aide
*R. H. Dean, ERDL, Phy.Sci. Aide
*C.E. Graham, ERDL, Phy. Sci. Aide
*T. W. Ostrom, Jr., MSC, U.S. Army

Research Participants

*E. Gloyna
*C. Smallwood, Jr.

Graduate Students for M.S. in San. Engr.

*** M. W. Carter
*** A. G. Friend
*** W. A. Hoyt
*** H. E. Butcher

Notes:

* No longer at ORNL
** AEC-TVA Ecological Survey of White Oak Creek - No longer under RLWRDS
*** Graduate student program completed

APPENDIX B

Oak Ridge Tap Water Analyses
(Four determinations)

M.O. Alkalinity (as $CaCO_3$)	90-99 ppm
Phenolphthalein Alkalinity (as $CaCO_3$)	0- 2 ppm
Soap Hardness (as $CaCO_3$)	90-108 ppm
Dissoved Solids	104-142 ppm
Non-volatile Solids	72- 80 ppm
Ca	23- 27 ppm
Mg	5- 8 ppm
Na	1- 6 ppm
SiO_2	2.5- 7 ppm
Nitrate	1.2- 1.75 ppm
pH	7.3- 7.9
Color	None
Odor	None
Dissolved CO_2	1- 3 ppm
Fe	< 0.03-0.04 ppm
Al	0.039- < 0.05 ppm
Cu	< 0.02- < 0.05 ppm
Ni	< 0.02- < 0.05 ppm
Cr	< 0.02- < 0.05 ppm
U	-- --
SO_4	9- 30 ppm
Cl	3.7-7.4 ppm
CO_3	54-59.4 ppm
HCO_3	112-120 ppm
PO_4	1.6-4.6 ppm
F	< 1.0-1.0

APPENDIX B

Oak Ridge Tap Water Analyses
(Four determinations)

M.O. Alkalinity (as $CaCO_3$)	90-99 ppm
Phenolphthalein Alkalinity (as $CaCO_3$)	0- 2 ppm
Soap Hardness (as $CaCO_3$)	90-108 ppm
Dissoved Solids	104-142 ppm
Non-volatile Solids	72- 80 ppm
Ca	23- 27 ppm
Mg	5- 8 ppm
Na	1- 6 ppm
SiO_2	2.5- 7 ppm
Nitrate	1.2- 1.75 ppm
pH	7.3- 7.9
Color	None
Odor	None
Dissolved CO_2	1- 3 ppm
Fe	$<$0.03-0.04 ppm
Al	0.039- $<$0.05 ppm
Cu	$<$0.02- $<$0.05 ppm
Ni	$<$0.02- $<$0.05 ppm
Cr	$<$0.02- $<$0.05 ppm
U	-- --
SO_4	9- 30 ppm
Cl	3.7-7.4 ppm
CO_3	54-59.4 ppm
HCO_3	112-120 ppm
PO_4	1.6-4.6 ppm
F	$<$1.0-1.0